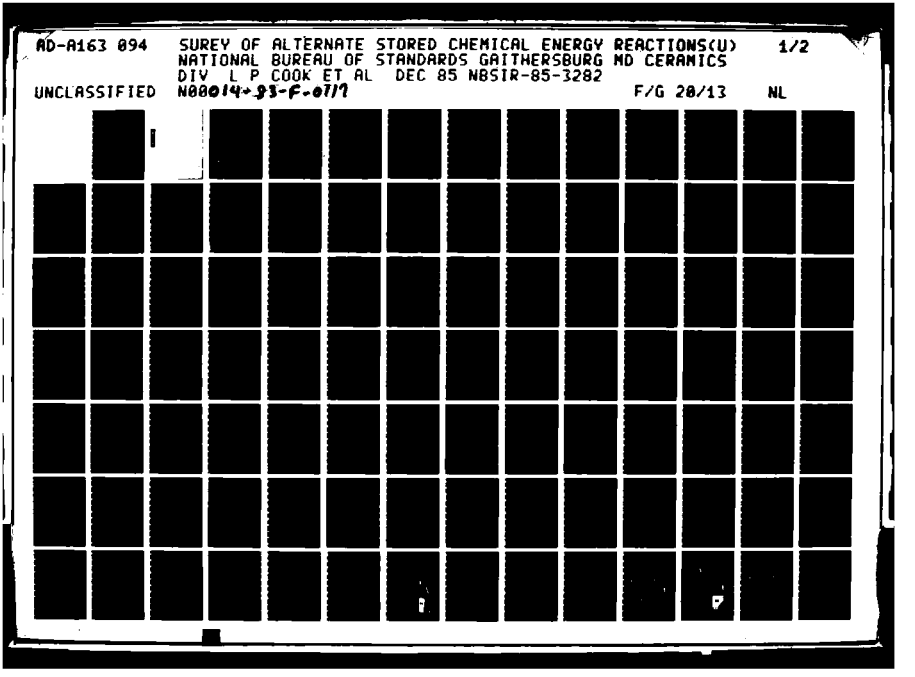
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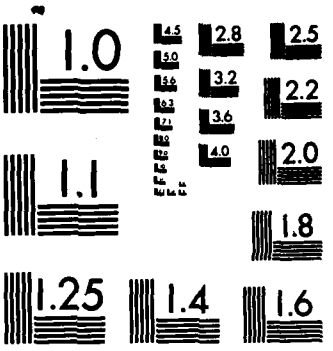

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SURVEY OF ALTERNATE STORED CHEMICAL ENERGY REACTIONS

L. P. Cook
E. R. Plante

U.S. DEPARTMENT OF COMMERCE
National Bureau of Standards
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December 1985

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Survey of Alternate Stored Chemical Energy Reactions

Annual Report

For the Period May 25, 1984 May 25, 1985

L. P. Cook and E. R. Plante

Inorganic Materials Division
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Survey of Alternate Stored Chemical Energy Reactions

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Abstract

A survey of eight alternative liquid metal stored chemical energy reactions has been made for purposes of comparison with the lithium-aluminum/water, lithium/sulfur hexafluoride, and other reaction schemes. The objective of the study was to survey the potential of these eight reactions as alternate stored chemical energy systems and to develop priorities for future study. Experimental data on the products of reaction and kinetics of reaction are presented for: Li/H_2O; H_2/O_2), (Li/H_2O; NaO_2/H_2O; H_2/O_2), ($MgAl/H_2O$; H_2/O_2), and $LiAl/ClO_3F$). These data have been collected using thermogravimetry and Knudsen effusion mass spectrometry, with x-ray diffraction analysis of experimental products. Among other results, the data show that the aluminum component of the fuels is relatively inert to oxidation up to 650 °C. Above this temperature, materials limitations have hampered the collection of experimental data.

Thermodynamic analysis has been used to extend the data on each of the eight reaction schemes, and to predict the chemical reaction which best represents the complete oxidation of each fuel by the indicated oxidant at 1100 K. Enthalpies have been calculated for each fuel/oxidant combination. Safety considerations are also discussed for each. Suggestions for future research are given, including suggestions for overcoming the materials problems encountered in this study.

I. Introduction

Recently there has been much interest in the use of light metals as fuel for stored chemical energy devices. A system currently in usage involves molten lithium metal and sulfur hexafluoride, according to the reaction,

$$8Li + SF_6 \rightarrow Li_2S + 6LiF \qquad [1]$$

Under development (1) is a system using lithium aluminum alloy as the fuel and water as the oxidant, according to the reaction:

$$LiAl + 2H_2O \rightarrow LiAlO_2 + 2H_2 \qquad [2a]$$

The hydrogen liberated is combined with oxygen to yield water:

$$2H_2 + O_2 \rightarrow H_2O \qquad [2b]$$

The water produced in this way is returned to [2a] to complete the cycle. In our laboratories investigation is continuing of reaction [2a], which is potentially much more complex than as written (2).

In addition to reaction [2], there are other possible alternate systems of interest and it is desirable to make a preliminary survey of these, to determine if certain ones might emerge as clear choices for further development, or at the least, if others might be eliminated from further consideration. The fuel/oxidant combinations that have been suggested are:

$Li/H_2O;\ H_2/O_2$ [3]

$Li/H_2O;\ NaO_2/H_2O;\ H_2/O_2$ [4]

$MgAl/H_2O;\ H_2/O_2$ [5]

$LiAl/ClO_3F$ [6]

$LiAlMg/ClO_3F$ [7]

$LiBe/ClO_3F$ [8]

$Li/C_{11}F_{20}$ [9]

LiB/NF_3 [10]

Combinations [3], [4], and [5] utilize a final stage of combustion in which hydrogen is burned, in a reaction identical to reaction [2b] above. Combinations [6] through [10], however, are envisioned as largely single-stage energy release schemes. To fully evaluate all these candidates it is desirable to have the following basic information:

(a) identity of the products of fuel/oxidant reaction

(b) an indication of the kinetics, especially the minimum temperature required for spontaneous combustion and self-sustaining reaction

(c) an estimate of the enthalpy of reaction

(d) knowledge of toxicity of reaction products

Information collected to date for each of the fuel/oxidant combinations is summarized in the paragraphs that follow. Most experiments were com-

pleted using a Mettler[1] thermoanalysis system, except for reaction [6], which was also investigated mass spectrometrically. Following a presentation of data for each of the above reaction schemes, a general discussion and recommendations for follow-up research are given. Results of a literature search are given in an Appendix (Section XIII).

II. Li/H_2O; H_2/O_2

A. Starting Materials and Container Materials

The starting material used for these experiments was finely powdered lithium metal (-140 mesh, 99.95% pure on a metal basis). This material had some surface oxidation upon receipt from the vendor, causing it to have a slightly grayish cast; the contaminants were tentatively identified as lithium carbide (Li_2C_2), lithium hydride, and lithium hydroxide, on the basis of weak x-ray diffraction peaks. All materials handling operations involving the lithium metal were completed in a glove box.

Quantitative study of the oxidation of metallic lithium poses special materials problems. Any container must be capable of withstanding attack by both metallic lithium and the oxidant. Silica glass is not a suitable container in the presence of both the lithium fuel and the water oxidant. Graphite and molybdenum are mentioned in the literature (3, 4) as container materials for lithium alloys; however both are attacked by oxidants at relatively low temperatures. A nickel-based alloy would appear to offer promise; however, nickel could

[1] Certain commercial equipment, instruments, or materials are identified in this paper in order to adequately specify the experimental procedure. Such identification does not imply recommendation or endorsement by the National Bureau of Standards, nor does it imply that the materials or equipment identified are necessarily the best available for the purpose.

be expected to undergo a certain degree of oxidation even at relatively low temperatures, affecting the results of sensitive thermogravimetric experiments, and possibly contaminating the sample. BN is a possible material, but experiments with LiAl (see below) suggest lithium abosrption. Perhaps the strongest candidate crucible materials for future work are BeO (5) and MgO; however, as these were not available, all measurements to date were completed using readily available alumina crucibles at temperatures below the melting point of lithium metal.

B. Reaction Products

Lithium metal was reacted with water-saturated flowing argon (30,000 ppm H_2O) in the thermoanalyzer. At 25 °C, slow reaction of lithium metal with water produces $LiOH.H_2O$. However the reaction probably proceeds sequentially, starting with LiOH, as evidenced by x-ray analysis of samples from different zones within the reaction cell. Near the bottom of the cell, products were largely LiOH; near the top, the hydrate was dominant. At 100 °C, the reaction product is almost entirely LiOH; small amounts of the hydrate are believed to have formed as the experiment cooled.

C. Kinetics

The finely powdered lithium metal used in these expeiments is highly reactive at room temperature in spite of the thin surface coating. This is evidenced by rapid reaction when deliberately exposed to the atmosphere. When placed in contact with water, violent reaction ensues, and explosion occurs due to spontaneous ignition of the evolved hydrogen.

In the thermoanalyzer experiments, the reaction was purposefully set up to proceed in controlled fashion. Reaction curves at 25 and 100 °C are shown in Fig. 1. The 25 °C curve shows a nearly linear segment from 0 to 1500 min. of reaction time, implying that reaction control in the early stages is not associated with build-up of a product layer, but rather may be linked to the rate of supply of the water vapor. The second half of the 25 °C curve is nonlinear. This could be due to the influence of the product layer of $LiOH.H_2O$, but is more likely to be due to the increased diffusion path required for vapor to reach the powders in the bottom of the cell. The reaction was stopped prior to completion. Theoretically completion would have occurred at a fractional weight gain of 5.1 (compared with 3.7 at the termination of the experiment). This number assumes complete conversion to $Li(OH).H_2O$.

By contrast, at 100 °C, the entire curve appears to be nonlinear. This suggests influence of the product layer (in this case LiOH). The reaction appears to have terminated in spite of the fact that the theoretical fractional weight gain of 2.5 was not reached. This probably indicates that the surface contamination of the Li powdered starting material is more severe than the x-ray analysis suggested. Future experiments will employ a much more nearly pure lithium powder.

D. Enthalpy

Thermodynamic data for calculation of the enthalpy of oxidation of molten lithium metal by H_2O are available in the JANAF tables (6). Enthalpy production in kcal per gram of fuel plus oxidant can be calculated, if the lithium oxidation reaction is combined with burning of the evolved hydrogen and recycling of water thus produced:

$$Li + H_2O \rightarrow LiOH + (1/2)H_2 \quad [11]$$

$$(1/2)H_2 + (1/4)O_2 \rightarrow (1/2)H_2O \quad [12]$$

$$Li + (1/2)H_2O + (1/4)O_2 \rightarrow LiOH \text{ (combined reaction)} \quad [13]$$

The calculated value[2] at 1100 K (827 °C) is 3.5 kcal/g. If LiOH did not form, the combined reaction would be:

$$2Li + (1/2)O_2 \rightarrow Li_2O \quad [14]$$

for which the enthalpy yield is 4.8 kcal/g. Clearly there is a heavy energy penalty paid by loss of hydrogen in the cycle to form LiOH.

E. Safety Considerations (7)

Lithium metal reacts exothermically with the moisture in human tissue, causing both thermal and chemical burns. An explosion hazard exists when the metal is exposed to moisture of any form, or to any other oxidizing material. When heated in air or burned it emits toxic fumes of Li_2O and LiOH. It reacts with nitrogen at high temperatures.

III. Li/H_2O; NaO_2/H_2O; H_2/O_2

A. Starting Materials

The Li/H_2O part of the reaction scheme has been discussed above. The role of NaO_2 is to provide oxygen for the second stage burning of hydrogen.

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[2]All energy yield calculations in this report are listed in terms of kcal per gram of fuel plus oxidant.

The NaO_2 used in these experiments was indicated by the vendor to be 95% NaO_2. X-ray powder diffraction analysis of the starting material which consisted of pale yellow granules, yielded a very complex pattern containing peaks for NaOH, NaO_2(I) and several unidentified peaks.

B. Reaction Products

NaO_2 was placed in a platinum cell in the thermoanalyzer. The sample was brought to temperature (25 or 100 °C) and then a stream of flowing water-saturated argon (30,000 ppm H_2O) was introduced. At the conclusion of the 100 °C experiment the sample was x-rayed. This was not possible for the 25 °C experiment, which produced liquid; apparently the NaO_2 (or the NaOH produced from it) is deliquescent at 25 °C. The 100 °C experiment yielded predominantly NaOH; the lesser amount of $NaOH.H_2O$ present is thought to have formed as the experiment cooled.

C. Kinetics

Reaction curves for NaO_2/H_2O at 25 and 100 °C are shown in Fig. 2. The 25 °C curve consists of two linear weight gain portions. Interestingly the slope increases for the later segment. This is interpreted as follows. During the early 25 °C segment, the reaction involves loss of oxygen and gain of water:

$$2NaO_2 + 3H_2O \rightarrow 2(NaOH.H_2O) + (3/2)O_2 \qquad [15]$$

The theoretical fractional weight gain for this reaction is 0.06, which is near the change in slope for the 25 °C curve. During the later segment of the 25 °C reaction curve in Fig. 2, the product is thought to be the deliquescent liquid observed at the conclusion of the experiment, produced according to the reaction:

$$NaOH.H_2O + H_2O \rightarrow (NaOH, H_2O)\ (\ell) \qquad [16]$$

Possibly formation of the liquid results in increased accessibility by the gas, explaining the increased slope.

At 100 °C, the reaction of NaO_2 with water vapor produces a nearly linear weight loss as would be expected from the reaction:

$$2NaO_2 + H_2O \rightarrow 2NaOH + (3/2)O_2 \qquad [17]$$

This reaction would yield a theoretical weight loss of 0.27. The 100 °C reaction apparently terminates at less than this value, reflecting the presence of the NaOH impurity noted in the x-ray analysis of the starting materials.

D. Enthalpy

If the sodium superoxide reaction is combined with the lithium/water oxidization reactions, we have the following:

$$(1/3)NaO_2 + (1/6)H_2O \rightarrow (1/3)NaOH + (1/4)O_2 \qquad [18]$$

$$Li + (1/2)H_2O + (1/4)O_2 \rightarrow LiOH \qquad [19]$$

$$(1/3)NaO_2 + (2/3)H_2O + Li \rightarrow (1/3)NaOH + LiOH \qquad [20]$$

(combined reaction)

An enthalpy of formation for NaO_2 at 298 K can be estimated as -62.7 kcal.

Neglecting the heat required to bring the oxygen from the first stage up to 1100 K, the total energy release in reaction [20] is -2.2 kcal/g. This is significantly less than the -3.5 kcal/g calculated earlier for the Li/H_2O; H_2/O_2 combination. However it may be that there are advantages associated with use of a solid as the oxidant source rather than a compressed gas.

E. Safety Considerations (7)

Hazards associated with lithium are noted in the discussion of Li/H_2O above. Sodium superoxide has much the same toxicity as NaOH, which it forms upon contact with moisture. Contact with skin could be expected to cause burns and ulceration; inhalation of the mist or powder could cause damage to the upper respiratory tract and lungs. Heating could cause violent decomposition. Contact with moisture or reducing agents could result in explosion.

IV. $MgAl/H_2O$; H_2/O_2

A. Starting Materials

Magnesium aluminum alloy of 50/50 atomic ratio was prepared in the NBS metals processing lab by melting high purity magnesium and aluminum together under a halide flux and casting the melt into a steel chill mold. The resulting ingot was very brittle and was easily crushed in a mortar and pestle in a glove box. A (-200, +400) sieve fraction was then prepared for the experiment and stored in the glove box until use. An x-ray diffraction pattern showed characteristic peaks for Al_2Mg_3 with a few additional unidentified peaks of low intensity.

B. Reaction Products

The powdered alloy was placed in an alumina cell in a glove gox and transferred to the thermoanalyzer system. Next the specimen was brought to 650 °C in an argon atmosphere. At this temperature the specimen was completely melted, according to the Mg-Al phase diagram (8). Next, argon saturated with water at 25 °C (30,000 ppm H_2O) was flowed continuously over the sample. At the conclusion of reaction the gray, porous sample was cooled, removed and x-rayed. The x-ray powder pattern showed strong peaks for aluminum metal and MgO (periclase). Apparently only the magnesium component of the molten alloy was oxidized, as there were no peaks for Al_2O_3, $MgAl_2O_4$ or any other aluminous phases.

C. Kinetics

Rate data for the oxidation of molten MgAl by water vapor are shown in Fig. 3. The meaning of the step in the weight gain curve at about 100 minutes of reaction time is not clear. However the two segments of this curve on either side of the break have substantially linear portions, suggesting that diffusion through a product layer of increasing thickness is not a controlling factor during most of the reaction. The reaction appears to slow to a very low rate as the fraction reacted approaches 0.30. This is in agreement with the x-ray analysis, which indicates essentially all the Mg has been oxidized; complete oxidation of the Mg to periclase would correspond to a fractional weight gain of 0.31, very close to that observed; on the other hand, complete oxidation of both the magnesium and the aluminum would result in a fractional weight gain of 0.62.

D. Enthalpy

Referring to the system Mg-Al-O-H, at least three possible oxidation reactions involving the 1:1 alloy and water could occurr. These are

summarized below:

$$2MgAl + 5H_2O \rightarrow MgO + MgAl_2O_4 + 5H_2 \quad [21a]$$

$$5H_2 + (5/2)O_2 \rightarrow 5H_2O \quad [21b]$$

$$2MgAl + 5H_2O \rightarrow 2MgO + Al_2O_3 + 5H_2 \quad [22a]$$

$$5H_2 + (5/2)O_2 \rightarrow 5H_2O \quad [22b]$$

$$MgAl + H_2O \rightarrow MgO + Al + H_2 \quad [23a]$$

$$H_2O + (1/2)O_2 \rightarrow H_2O \quad [23b]$$

To complete enthalpy calculations for these reactions an estimate for the enthalpy of formation of molten MgAl was made using the ideal solution approximation. Calculated enthalpy release values for the three reaction pairs are respectively, -3.8, -3.8, and -2.2 kcal/g. Clearly, formation of spinel (reaction [21a] vs. reaction [22a]) affects the energy yield very little. Reaction [23a] has been included because it is the one observed, and clearly its occurrence instead of [21a] or [22a] results in substantially lower energy yield. Presumably at some higher temperature the aluminum component will begin to react according to reaction [21a] or [22a].

E. Safety Considerations (7)

Hazards data are not available for MgAl alloy. Aluminum is not regarded as a poison, although inhalation of finely divided powders can damage the lungs. Magnesium on the other hand can cause chemical gas

gangrene if it enters the body, due to generation of hydrogen. Fine magnesium powder readily ignites by exposure to spark or flame.

V. LiAl/ClO_3F

A. Starting Materials

The starting material for these experiments was powdered LiAl. The material as received was packed under argon and visually showed no signs of surface contamination--all surfaces were bright and shiny. X-ray analysis showed only peaks of LiAl. All manipulations involving this material were done in a glove gox through which a continuously purified argon atmosphere was recirculated. The material used in these experiments was a (-200, +400) sieve fraction. Transfer to the thermoanalyzer was accomplished in a glove bag, flushed repeatedly with purified argon.

The ClO_3F (perchloryl fluoride) was obtained from the vendor in cylinders containing either 100 g or 250 g. Principal impurities were N_2, O_2, CO_2, H_2O and HF (see section on mass spectrometry).

Gas was run through a corrosive gas regulator and then through a column containing NaOH pellets and molecular sieve, and finally through a flowmeter and then into the thermoanalyzer. The ClO_3F, a very dense gas, was piped directly into the crucibles in the thermoanalyzer experiments. Provision was made for mixing the ClO_3F with continuously variable amounts of argon. However, all thermoanalyzer experiments reported were carried out using a flow of pure ClO_3F since it was discovered that the reactions proceeded in controlled fashion so that no argon dilution was necessary. Although argon was used to flush the balance components

during the experiments, it is estimated that the effective partial pressure of ClO_3F over the specimens during these experiments was close to 1 atmosphere.

B. Reaction Products

Initially, experiments were conducted in alumina crucibles; however these were unsatisfactory above 700 °C, owing to the corrosiveness of the molten alloy. Crucibles of $LiAlO_2$ have been fabricated and these appear to offer promise for application in higher temperature experiments. Additionally MgO and BeO offer promise. Boron nitride crucibles were fabricated and tested in contact with LiAl at 800 °C. They seem to hold up to attack by the molten alloy well, except for a slight discoloration, which may be associated with diffusion of Li. Also a steady and extensive weight gain of the crucible occurred during preliminary tests under an atmosphere of purified argon. As we have no explanation for or solution to this problem, use of the BN crucibles was discontinued. Calcia-stabilized zirconia and silica glass crucibles were employed for the mass spectrometric studies, with mixed results. All other experiments reported employed alumina crucibles.

At the conclusion of the LiAl/ClO_3F experiments, crucibles were removed and placed in an inert atmosphere. X-ray mounts were prepared in the glove box and x-rayed under flowing N_2. Results of x-ray analysis are indicated in Table 1, below.

As can be seen from Table 1, experimental results vary greatly depending upon experimental conditions. The following generalizations can be made, however, for the range of conditions investigated (350-650 °C, P_{ClO_3F} = 1 atm; or, 900 °C, P_{ClO_3F} = 10^{-5} atm).

(1) Fluorides (Li_3AlF_6, AlF_3, LiF) appear to be by far the dominant reaction products.

(2) Chlorides may form during initial stages of oxidation[3] (see discussion of kinetics) but are relatively minor phases in the end products.

(3) The lithium in the alloy is preferentially oxidized leaving behind aluminum which behaves as though inert (see also discussion of MgAl oxidation).

(4) The occurrence of oxides as oxidation products is limited in these experiments to low partial pressures of ClO_3F and/or high temperatures.

C. Mass Spectrometry

i) Outline of Method

In this technique, the Li-Al alloy contained in a Knudsen cell is exposed to ClO_3F introduced into the cell through a gas inlet system. At appropriate temperatures, reaction products undergo vaporization into the source of the mass spectrometer where the gaseous products are ionized and mass analyzed. The major advantage of this method is that it can in principle detect gaseous reaction products. A second advantage is that the number of collisions of reactant vapor with the condensed phase may be controlled at a low value so that the kinetics of the process can be substantially solved.

In the present measurements, however, it was found that reaction rates are quite slow even up to temperatures as high as 1000 °C so that modifications in the experimental arrangement to enhance reaction rates will be advantageous.

[3]Oxidation is here used in the generic chemical sense.

ii) Vaporization Considerations

At low temperatures some of the reaction products may be relatively non-volatile so that they will not be detected by mass spectrometric analysis. On the basis of data in the literature, the metal chlorides are more volatile than the metal fluorides while the metal oxides are non-volatile up to temperatures of 1000 °C or so. Also, because the halides will form a salt solution, complexing to form interhalide compounds may influence the volatity of the solution. For example, formation of $LiAlCl_4$ would greatly reduce the pressure expected from formation of $AlCl_3$.

iii) Experimental set-up

These measurements were made using a fused silica effusion sampler. The Li-Al alloy was contained in a calcia-stabilized zirconia cup supported within the effusion sampler. The ClO_3F was delivered to the gas inlet tube through a small leak valve. The Li-Al alloy was loaded into the zirconia cell in a dry-box then transferred to the mass spectrometer under argon to reduce the reaction of the Li-Al alloy with ambient water vapor.

An effusion orifice of 1 mm diameter was used in the effusion sampler. This proved to be too large as the ClO_3F reacted much less rapidly with the Li-Al alloy than expected.

The control valve was attached to the ClO_3F cylinder in the dry box so the ClO_3F was diluted with Ar during the earlier parts of the experiment.

iv) Analysis of ClO_3F

Mass spectrometric sampling of the commercial ClO_3F gas showed predominant peaks at m/e values of 104, 102, 85, 83, 69, 67, 53, and 51 corresponding to the ^{37}Cl and ^{35}Cl isotopes of the species ClO_3F^+,

ClO_3^+, ClO_2^+, and ClO^+. Except for ClO_3F^+, these peaks are due to fragmentation of the ClO_3F during the ionization process using electrons at 30v. Besides the ClO_3F, Ar which was introduced from the dry box installation of the regulator was also observed. By comparing the mass spectrum with the ClO_3F gas flow on or off, it was determined that the ClO_3F also contained a small amount (perhaps a percent or so) of air (N_2, O_2, CO_2, and H_2O). The H_2O had apparently partially reacted with the steel cylinder and produced slight traces of HF (mass 20), $H^{35}Cl$ (36), and $H^{37}Cl$ (38). However, it is not likely that the contamination had significant effect on the reaction of ClO_3F with the alloy.

v) Reaction of Li-Al Alloy with ClO_3F

The first mass peaks due to reaction of ClO_3F with the Li-Al alloy were observed at 420 °C. These peaks were Li^+ from fragmentation of LiCl(g), $LiCl^+$ from ionization of the LiCl(g) and Li_2Cl^+, from ionization of the Li_2Cl_2(g) parent.

One might expect Al-Cl species in this temperature range but none were observed, in spite of the fact that loss of volatile substances is indicated by the analogous thermogravimetric experiment. It is clear from the presence of both LiCl(g) and Li_2Cl_2(g) that a condensed halide phase was formed on the sampler.

With time, the ratio of $LiCl^+/Li_2Cl^+$ gradually increased. This probably showed that LiF was also forming in the condensed phase and reducing the activity of LiCl. At 600 °C, Li_2F^+ from the fragmentation of Li_2F_2 was observed in the mass spectrum. Over the temperature range from 600-900 °C ions corresponding to parent molecules LiCl, Li_2Cl_2, LiF, and Li_2F_2 were observed. It did not appear that any Al bearing species were formed in the gas phase.

At the conclusion of the experiment, the Li-Al alloy still had a powdery appearance even through it had been heated several hundred degrees above the melting point.

An x-ray diffraction pattern of the residue revealed the presence of $LiAlO_2$ with perhaps an equal amount of unreacted Al (see Table 1).

vi) Al-ClO_3F Interaction

Because Al bearing species were not observed from the Li-Al alloy, a mass spectrometric experiment was carried out using only powdered Al in the effusion cell. The purpose of this experiment was to see if it was possible to form Al bearing species by reactions with ClO_3F. Near 500 °C species corresponding to the halide ions AlF^+, $AlCl^+$, AlF_2^+, $AlClF^+$, Al_3^+, AlF_2Cl^+, $AlCl_2F^+$, and $AlCl_3^+$ were observed. Because of the number of complex species in the vapor phase, the existence of a condensed phase at this temperature consisting of mainly AlF_3 seems likely. The duration of the experiment during which Al-halide species was generated was quite short. All of the Al bearing species decreased with time and increasing the ClO_3F flow rate and the temperature had no effect on increasing their intensity. This seems to indicate that the chlorination and fluoridation processes took place at only selected sites in the Al powder which were gradually depleted or lost, perhaps because of oxidation.

At 760 °C, most of the Al halide peaks had vanished except for AlF_2^+ which is a major fragmentation product of $AlF_3(g)$. The experiment was terminated after a brief heating period at 800 °C. Visual examination of the residue at the conclusion of the experiment revealed that most of the sample still had a powdery texture but a few small spheres resulting from melting the sample were also present. Since the sample

was heated to about 150 °C above the melting point of Al, the powdery texture of the sample must result from an oxide skeleton on the outside surface of each grain in the powder. The sample initially weighed 118 mg and gained 2.2 mg during the experiment, despite loss of the halides, indicating that a small amount of oxidation took place.

D. Kinetics

Thermogravimetric rate data for the reaction of LiAl with ClO_3F are shown in Fig. 4 at temperatures of 350, 450, 550 and 650 °C. It is presumed on the basis of the Li-Al phase diagram (9) that the LiAl was solid during these experiments. With the exception of the 350 °C experiment, rate curves all initially show a rapid weight gain at a nearly steady rate. This continues until a weight fraction gain of between 0.5 and 0.7 has been reached. At this point the rate of weight gain decreases for a short time and then increases again. This behavior is especially pronounced in the 650 °C experiment, where a small weight loss with time is apparent, followed by an increase in the rate of weight gain. This weight loss (and possibly, as well, the temporary decrease in rate of gain at all four temperatures) is best interpreted as due to formation of a volatile reaction product, most probably $AlCl_3$. Continued reaction leads to a similar ultimate weight gain of between 0.9 and 1.1 for experiments at all four temperatures, 350, 450, 550 and 560 °C (see Table 1). This is true in spite of the fact that the products are somewhat different in each case. Table 2 lists hypothetical reactions of LiAl with Cl, O and F and combinations thereof, together with theoretical weight gains. These reactions have been derived with the aid of thermodynamically calculated equilibrium phase compatibilities shown in Fig. 7. Calculated weight gains are highly variable, but the partial fluorination reactions [26] and [27] of Table 2 best approximate the

experimental products observed (Table 1). The fractional weight gains calculated for these reactions are 0.56 and 1.12, respectively, encompassing the range observed.

It seems likely from the curves in Fig. 4 that more than one reaction has occurred during the oxidation of LiAl by ClO_3F. It also seems likely that early stage condensed phase products undergo subsequent reaction with time to yield a new generation of products. In future experiments, reacion of LiAl with ClO_3F will be completed at higher temperatures, where hopefully a rapid approach to equilibrium will eliminate many of these complications. It is hoped that mass spectrometer measurements will be carried out at higher pressures of ClO_3F using a transpiration system.

E. Enthalpy

From Table 1 it can be seen that the reaction products at temperatures of 650 °C and below differ greatly, depending upon temperature. Further, oxidation of the alloy is far from complete, and the majority of the aluminum component of the alloy appears to have remained in the metallic state. The reaction products suggest disequilibrium among themselves; for example, Li_3AlF_6, AlF_3 and LiF are not in stable equilibrium together. The principal difficulty then is in extrapolating these limited results to a higher temperature such as 1100 K, at which oxidation could be expected to be much more complete, so that a meaningful enthalpy yield calculation can be done.

The LiAl/ClO_3F reaction can be described by the five component chemical system Li-Al-Cl-O-F. Fortunately thermodynamic data are available for many of the end-member compounds in this system and so a preliminary thermodynamic analysis can be completed. This results in an

estimated equilibrium reaction for complete oxidation which can be used to calculate enthalpy yield. The thermodynamic analysis was completed as follows. Fig. 5 depicts graphically the compositional regions of interest in the three principal quaternary subsystems contained within the five component system. Fig. 6 shows calculated phase compatibilities for important ternary reciprocal systems in Fig. 5. These compatibilities are schematic and indicate major features of phase equilibria which are consistent with known thermodynamic data, according to the reactions in Table 3. On the basis of Fig. 6, Fig. 7 has been constructed. This shows calculated equilibrium relations at 1100 K for the phase regions shown in Fig. 5. Based on Fig. 6 and 7, Fig. 8 has been constructed, showing calculated equilibrium relations at 1100 K among phases most likely to form as products of complete reaction of LiAl and ClO_3F. The reaction suggested by Fig. 8 is:

$$2LiAl + ClO_3F \rightarrow Al_2O_3 + LiF + LiCl \qquad [74x]$$

It should be emphasized that there is no direct experimental evidence yet in support of this reaction: however on purely thermodynamic grounds it is the favored reaction. To complete the enthalpy calculation, data for LiAl were estimated using the ideal solution approximation. The resulting value of enthalpy yield for this reaction is 3.8 kcal/g.

F. Safety Considerations (7)

Safety hazards associated with LiAl are similar to those associated with Li as noted above, although reaction with moisture is considerably less vigorous at room temperature. Perchloryl fluoride is a highly toxic gas which causes symptoms of asphyxiation and poisoning similar to

CO, although recovery is said to be rapid provided exposure has not been prolonged. The gas has a sweet odor, but this cannot be relied upon to indicate toxic concentrations. Perchloryl fluoride is readily absorbed through the skin. Although nonflammable, it does support combustion, as it is a powerful oxidizer. When heated to decomposition, it emits highly toxic fumes of chlorides and fluorides.

VI. LiAlMg/ClO_3F

A. Enthalpy

At the time of writing, experiments have not yet been completed in this system, owing to lack of available starting material. Therefore enthalpy yield estimates must be made solely on the basis of preliminary thermodynamic analysis. The fuel/oxidant reaction is represented by the 6-component system Li-Al-Mg-Cl-O-F. The phase equilibrium topology is of course impossible to represent in three dimensions; nonetheless, progress can be made by consideration of the six quaternary reciprocal systems shown in Fig. 9. By calculation of the reactions in Table 4, the schematic ternary phase compatibilities shown in Fig. 10 have been derived. From additional reactions in Table 4 and compatibilities in Fig. 10, construction of Figs. 11 and 12 was made possible. Fig. 11 pertains to progressive chlorination, fluorination or oxygenation of fuel. Fig. 12 relates to the equilibrium products of complete reaction. Table 5 gives some postulated equilibrium reactions involving progressive reaction of LiAlMg fuel with Cl, F, O and combinations thereof.

Based on the above considerations, the following is thought to represent complete reaction of LiAlMg alloy (1:1:1 atomic) with ClO_3F:

$$4LiAlMg + 3ClO_3F \rightarrow MgF_2 + LiF + 3LiCl + 2MgAl_2O_4 + MgO \qquad [134]$$

The calculated enthalpy yield for this reaction at 1100 K is 3.6 kcal/g.

B. Safety Considerations (7)

Safety hazards are similar to those noted for $LiAlMg/ClO_3F$ above.

VII. $LiBe/ClO_3F$

A. Enthalpy

Because of the toxicity of beryllium and beryllium compounds, experiments have not been conducted on the oxidation of lithium-beryllium alloy by ClO_3F. Hence any estimate of enthalpy release must rely soley upon thermodynamic estimates of the actual reaction. A thermodynamic analysis of the five component system Li-Be-Cl-O-F was completed as follows. Fig. 13 shows the three quaternary subsystems of interest. Based on reactions in Table 6, the ternary phase compatibilities in Fig. 14 have been constructed. Based on these compatibilities and additional reactions in Table 6, the schematic quaternary phase compatibilities in Fig. 15 have been derived; these show the results of progressive chlorination, fluorination, or oxygenation of LiBe fuel (Table 7 lists equilibrium reactions involving LiBe fuel with various combinations of Cl, O and F). Fig. 16 indicates calculated compatibilities among likely reaction products. The reaction postulated on the basis of thermodynamic data for complete oxidation of LiBe by PCF is:

$$8LiBe + 3ClO_3F \rightarrow 8BeO + Li_2O + 3LiF + 3LiCl \qquad [169]$$

The enthalpy relased by this reaction at 1100 K would be 4.6 kcal/g.

B. Safety Considerations (7)

Lithium and beryllium do not form stable intermetallic compounds together (10) and would be expected to be present together in fuel as the chemically uncombined metals. For a discussion of Li safety hazards, see Li/H_2O above. Beryllium is highly toxic both in metallic form and in compound form. Many of the beryllium salts have high vapor pressures, especially when heated. Symptoms of exposure usually involve respiratory distress of varying severity; symptoms may be delayed for several years after exposure. Chronic berylliosis can lead to death from respiratory and cardiac failure. In addition to harm incurred by inhalation, beryllium and its compounds also act on the skin, causing lesions and tumors. Beryllium by itself does not present an explosion hazard, however the admixed lithium in the fuel obviously does. For a discussion of perchloryl fluoride safety refer to $LiAl/ClO_3F$ above.

VIII. $Li/C_{11}F_{20}$

A. Starting Materials

The fluorocarbon compound $C_{11}F_{20}$ was not obtainable from commercial suppliers; the closest molecular weight compound available was the structurally similar double ring compound $C_{11}F_{19}$, an apparently stable, clear liquid. Experiments have not yet been completed due to the necessity of installing a special furnace with a boiling chamber for the liquid oxidant on the thermoanalyzer. It is hoped that loan from another facility of a water vapor furnace (which would also provide 1 atm of P_{H_2O} for the Li/H_2O and $MgAl/H_2O$ experiments) will provide a solution to this experimental problem.

B. Enthalpy

In the absence of experimental data, it is again necessary to utilize thermodynamic estimates in arriving at an oxidation reaction for the enthalpy calculation. The system Li-F-C provides a compositional representation of the $Li/C_{11}F_{20}$ reaction. By using calculated reactions in Table 8, Fig. 17 can be constructed, showing schematic phase compatibilities at 1100 K. On this basis, the thermodynamically estimated oxidation reaction is:

$$C_{11}F_{20} + 20Li \rightarrow 11C + 20LiF \qquad [176]$$

As no enthalpy data have been located for $C_{11}F_{20}$, it is necessary to make an estimate. This can be done by extending the procedures used for estimating properties of hydrocarbon molecules. A very approximate value of 900 kcal/mole is obtained in this way for 298 K. The calculated enthalpy yield at 298 K is 3.1 kcal/g. Presumably the value at 1100 K would be somewhat higher.

C. Safety Considerations (7)

For a discussion of Li safety hazards, see Li/H_2O above. Data are not available for the $C_{11}F_{19}$ obtained, but the related compound $C_{11}F_{20}$ is a highly stable molecule which has been used to develop a synthetic blood substitute.

IX. LiB/NF_3

A. Starting Materials

NF_3 has been obtained from a commercial source in small cylinders.

Principal impurities are stated by the vendor as N_2, HF, NO and H_2O. A source of LiB has been located but the material has not yet been procured, and so there are not experimental data on the reaction at this point in time.

B. Enthalpy

In the absence of experimental data on the LiB/NF_3 reaction, thermodynamic analysis of the system Li-B-N-F (Fig. 18) is essential. Using the reactions in Table 9, the schematic 1100 K ternary phase relations in Fig. 19 have been calculated. Fig. 20 gives calculated compatibilities among product phases at 1100 K. Table 10 gives reactions for progressive degrees of oxidation of LiB fuel by NF_3. The suggested reaction for complete oxidation of LiB by NF_3 is:

$$3LiB + 2NF_3 \rightarrow 3LiF + BN \qquad [185]$$

Boron trifluoride (BF_3) is a gas even at relatively low temperatures, and so this would not be a desirable reaction. However by simply adding excess lithium to the fuel, in principle the following reaction could be obtained:

$$2Li + LiB + NF_3 \rightarrow 3LiF + BN \qquad [186]$$

Depending upon the kinetics, BF_3 might still form as an intermediate product, but the equilibrium final partial pressure would be very low. Although LiB is known as a stable compound, thermochemical data were not available. It has been assumed that at 1100 K it would be molten, and the ideal solution approximation has been applied to give an enthalpy of formation of 0.0 kcal. The enthalpy yield of reaction [180] can then be calculated as 4.6 kcal/g.

C. Safety Considerations (7)

Handling properties and toxicity of the compound LiB apparently

have not found their way into appropriate compendiums, but it is reasonable to assume that the comments made above under discussion of safety for Li and LiAl fuels would also apply here. The compound is used in pyrotechnical devices, and so there may be a potential explosion hazard. With regard to NF_3, it is a colorless gas with a moldy odor and a high toxicity similar to HF. It could therefore be expected to be extremely corrosive to the respiratory tract and skin. Prolonged exposure may cause skeletal changes.

X. Summary and Discussion

In terms of energy release/unit mass, ranking of various stored energy reactions is as in Table 11, which includes for reference, two schemes not otherwise discussed in this report:

$$Mg + H_2O \rightarrow MgO + H_2 \qquad [187a]$$

$$H_2 + (1/2)O_2 \rightarrow H_2O \qquad [187b]$$

$$2Al + 3H_2O \rightarrow Al_2O_3 + 3H_2 \qquad [188a]$$

$$3H_2 + (3/2)O_2 \rightarrow 3H_2O \qquad [188b]$$

Several points should be made regarding the ranking in Table 11. First, direct experimental evidence for several of these reactions has not yet been obtained, due in part to materials limitations. Although we are optimistic that these will soon be overcome, in the meantime the ranking

in Table 11 may have only limited practical significance. An example is the reaction Li/H_2O [14]. Theoretically it should be high on the list at 4.8 kcal/g. However in practice this yield may not be achieved due to formation of LiOH, which irreversibly ties up hydrogen in the water cycle, and reaction [13], with an enthalpy yield of 3.5 could prevail instead. Also there are presumably other factors such as safety to be considered. The high toxicity of beryllium makes the practicality of utilizing reaction [169] questionable. On the other hand, it could probably be said that since fuel or oxidant or both are inherently chemically unstable, all of the reaction schemes involve some degree of toxicity to human tissue or other safety hazards, and suitable safeguards must be employed with any reaction scheme to eliminate the possibility of containment failure or runaway reaction. For example, lithium metal exposed to moist air at room temperature rapidly generates hydrogen. Perchloryl fluoride has a toxic effect similar to CO. NF_3 is not only toxic but extremely hazardous under pressure. When all such factors are considered, undoubtedly the Mg/H_2O, $MgAl/H_2O$ and Al/H_2O schemes pose the least safety hazard; however the latter two fuels may require very high temperature operation to overcome the kinetic barriers to oxidation of the aluminum. In terms of safety the $LiAl/H_2O$ scheme is also high on the list, as LiAl is much less reactive than lithium upon exposure to air. Furthermore from a practical viewpoint, there may be other factors to consider besides enthalpy yield. For example, although the $Li/C_{11}F_{20}$ and Li/H_2O; NaO_2/H_2O schemes rank low in Table 11, there may be certain advantages associated with not having to store gaseous oxidizers.

Additional experimental work is necessary at higher temperatures than attained to date in order to verify the reactions presented in

Table 11. Evidence to date suggests several of the reactions are kinetically hindered at temperatures below 650 °C. We are optimistic that the materials limitations encoutered in this preliminary study will be overcome, so that relevant experimental data on high temperature kinetics and phase equilibria can be systematically gathered in the near future.

XI. Future Work

The highest priority involves finding suitable container materials so that thermogravimetric experiments can be extended to higher temperatures. Rod stock of high density MgO has been obtained and is being fabricated into crucibles. Also beryllia crucibles will be machined to order by a vendor equipped to do this safely. Crucibles of $LiAlO_2$ have already been fabricated. We are optimistic that use of one or more of these materials will allow us to extend the temperature range of experimentation into the 650-1000 °C region.

After the container problem is solved the first priority will be to determine if the oxidation reactions can be made to go to completion, and if so what the minimum temperature is at which this will occur. Once optimum conditions for reaction are determined, an attempt will be made to measure enthalpy of reaction directly using DTA.

Simultaneously with these experiments phase equilibrium measurements will commence. The first experiments will involve heating the predicted end products of the reactions together in sealed capsules, quenching and examining to see if they really are in equilibrium as suggested by the thermodynamically calculated reaction. This will give important information on solubilities (both liquid and solid) as well.

It is hoped that mass spectrometric measurement of species vapor pressures during these reactions can be completed using the NBS transpiration mass spectrometry system. This apparatus allows measurements of species at pressures up to 10 atm, which would more closely simulate actual reactor conditions. Top priority would be given to the investigation of the LiAl/ClO_3F and LiAlMg/ClO_3F systems. Also, the (Li + LiB)/NF_3 and Li/$C_{11}F_{20}$ systems might be amenable to investigation by this approach.

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XIII. Appendix: Results of Literature Survey

Following are bibliographic listings which have been selected from a computerized literature search of the Chemical Abstracts Data Base (1967-1984). References are not annotated, and low temperature as well as high temperature studies have been included. References are loosely grouped according to search categories that roughly correspond to the various alternate stored chemical energy reaction fuels and oxidants.

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XIV. Tables

Table 1. X-ray Diffraction Analysis of $LiAl/ClO_3F$ Experimental Products

Expt.	Max Temp.	%Wt. Gain	Phases
TGA	650 °C (isothermal)	109.86	α-Li_3AlF_6, LiF*, AlF_3, Al
TGA	550 °C (isothermal)	97.58	Al, β-Li_3AlF_6, AlF_3, LiF
TGA	450 °C (isothermal)	98.52	Al, β-Li_3AlF_6, AlF_3, LiF, (LiCl)
TGA	350 °C (isothermal)	109.26	Al, AlF_3, Li_3AlF_6,
Mass Spec.	900 °C (non-isothermal)	---	$\gamma LiAlO_2$, Al
TGA	650 °C (non-isothermal)	91.69	αLi_3AlF_6, AlF_3, Al, LiF
TGA	650 °C Partial React. (isothermal)	50.43	LiCl, Al, LiAl, $\gamma LiAlO_2$, LiF

*LiF apparently contains a small amount LiCl in solid solution, as judged by x-ray line shifts

Table 2. Progressive Oxidation of LiAl alloy by Reaction with Components of ClO_3F

Reaction #	Type of Reaction	Reaction	Final Wt. Gain at Completion
[24]	chlorination	$LiAl + ClO_3F \rightarrow LiCl + Al + (3/2)O_2 + (1/2)F_2$	+0.51
[25]	chlorination	$LiAl + 4ClO_3F \rightarrow LiCl + AlCl_3(g) + 6O_2 + 2F_2$	+0.25
[26]	fluorination	$LiAl + ClO_3F \rightarrow LiF + Al + (3/2)O_2 + (1/2)Cl_2$	+0.56
[27]	fluorination	$3LiAl + 6ClO_3F \rightarrow Li_3AlF_6 + 2Al + 9O_2 + 3Cl_2$	+1.12
[28]	fluorination	$3LiAl + 12ClO_3F \rightarrow Li_3AlF_6 + 2AlF_3 + 18O_2 + 6F_2$	+2.24
[29]	oxygenation	$3LiAl + 2ClO_3F \rightarrow 3LiAlO_2 + Cl_2 + F_2$	+0.94
[30]	chlorination & oxygenation	$5LiAl + 2ClO_3F \rightarrow 3LiAlO_2 + 2LiCl + 2Al + F_2$	+0.98
[31]	chlorination & oxygenation	$7LiAl + 4ClO_3F \rightarrow 3LiAlO_2 + 4LiCl + 2Al_2O_3 + 2F_2$	+1.41
[32]	fluorination & oxygenation	$5LiAl + 2ClO_3F \rightarrow 3LiAlO_2 + 2LiF + 2Al + Cl_2$	+0.79
[33]	fluorination & oxygenation	$7LiAl + 4ClO_3F \rightarrow 3LiAlO_2 + 4LiF + 2Al_2O_3 + Cl_2$	+1.12
[34]	chlorination & fluorination	$2LiAl + ClO_3F \rightarrow LiCl + LiF + 2Al + (3/2)O_2$	+0.80
[35]	chlorination & fluorination	$9LiAl + 6ClO_3F \rightarrow 6LiCl + Li_3AlF_6 + 8Al + 9O_2$	+1.07
[36]	chlorination & fluorination	$3LiAl + 6ClO_3F \rightarrow 3LiCl + 2AlF_3 + AlCl_3(g) + 9O_2$	+1.90
[37]	chlorination & fluorination & oxygenation	$7LiAl + 2ClO_3F \rightarrow 2LiCl + 3LiAlO_2 + 2LiF + 4Al$	+0.86
[38]	chlorination & fluorination & oxygenation	$2LiAl + ClO_3F \rightarrow Al_2O_3 + LiF + LiCl$	+1.51

Table 3. Calculated Phase Compatibilities in System Li-Al-O-Cl-F at 1100 K

Reaction #	Subsystem	Reaction	ΔG_R(1100 K), kcal
[39]	Li_2O-Al_2O_3-Li-Al	$2Li_2O(c) + LiAl(\ell)$ $LiAlO_2(c) + 4Li(\ell)$	- 9.6
[40]	Li_2O-Al_2O_3-Li-Al	$2Li_2O(c) + Al(\ell) \rightarrow LiAlO_2(c) + 3Li(\ell)$	- 11.1
[41]	Li_2O-Al_2O_3-Li-Al	$3Li(\ell) + 2Al_2O_3(c) \rightarrow 3LiAlO_2(c) + Al(\ell)$	- 47.6
[42]	Li_2O-Al_2O_3-Li-Al	$3LiAlO_2(c) + 2Al(\ell) \rightarrow 2Al_2O_3(c) + 3LiAl(\ell)$	+ 43.1
[43]	Li_2O-Al_2O_3-Li-Al	$3Li_2O(c) + 2LiAl(\ell) \rightarrow 8Li(\ell) + Al_2O_3(c)$	+ 10.1
[44]	Li_2O-Al_2O_3-Li-Al	$Al_2O_3(c) + 6LiAl(\ell) \rightarrow 3Li_2O(c) + 8Al(\ell)$	+ 2.0
[45]	Li_2O-Al_2O_3-Li-Al	$LiAlO_2(c) + 3LiAl(\ell) \rightarrow 2Li_2O(c) + 4Al(\ell)$	+ 15.7
[46]	Li_2O-Al_2O_3-Li-Al	$3LiAlO_2(c) + LiAl(\ell) \rightarrow 2Al_2O_3(c) + 4Li(\ell)$	+ 49.2
[47]	Li_2O-Al_2O_3-Li-Al	$3Li_2O(c) + 2Al(\ell) \rightarrow Al_2O_3(c) + 6Li(\ell)$	+ 7.1
[48]	Li_2O-Al_2O_3-LiCl-$AlCl_3$	$2Li_2O(c) + AlCl_3(g) \rightarrow LiAlO_2(c) + 3LiCl(\ell)$	-117.5
[49]	Li_2O-Al_2O_3-LiCl-$AlCl_3$	$6LiCl(\ell) + Al_2O_3(c) \rightarrow 3Li_2O(c) + 2AlCl_3(g)$	+205.6
[50]	Li_2O-Al_2O_3-LiCl-$AlCl_3$	$3LiCl(\ell) + 2Al_2O_3(c) \rightarrow 3LiAlO_2(c) + AlCl_3(g)$	+ 58.7
[51]	LiCl-$AlCl_3$-LiF-AlF_3	$3LiCl(\ell) + AlF_3(c) \rightarrow 3LiF(c) + AlCl_3(g)$	+ 32.7
[52]	LiCl-$AlCl_3$-LiF-AlF_3	$3LiCl(\ell) + Li_3AlF_6(\ell) \rightarrow 6LiF(c) + AlCl_3(g)$	+ 44.1
[53]	LiCl-$AlCl_3$-LiF-AlF_3	$Li_3AlF_6(\ell) + AlCl_3(g) \rightarrow 3LiCl(\ell) + 2AlF_3(c)$	- 21.4
[54]	LiCl-$AlCl_3$-Li-Al	$3LiAl(\ell) + AlCl_3(g) \rightarrow 3LiCl(\ell) + 4Al(\ell)$	-101.8
[55]	LiCl-$AlCl_3$-Li-Al	$4Li(\ell) + AlCl_3(g) \rightarrow 3LiCl(\ell) + LiAl(\ell)$	-107.9
[56]	LiCl-$AlCl_3$-Li-Al	$3LiCl(\ell) + Al(\ell) \rightarrow 3Li(\ell) + AlCl_3(g)$	+106.4

Table 3 (continued)

Reaction #	Subsystem	Reaction	ΔG_R(1100 K), kcal
[57]	Li_2O-Al_2O_3-LiF-AlF_3	$6LiF(c) + Al_2O_3(c) \rightarrow 3Li_2O(c) + 2AlF_3(c)$	+140.2
[58]	Li_2O-Al_2O_3-LiF-AlF_3	$LiAlO_2(c) + Li_3AlF_6(\ell) \rightarrow 2Li_2O(c) + 2AlF_3(c)$	+ 96.1
[59]	Li_2O-Al_2O_3-LiF-AlF_3	$3LiAlO_2(c) + Li_3AlF_6(\ell) \rightarrow 6LiF(c) + 2Al_2O_3(c)$	- 14.7
[60]	Li_2O-Al_2O_3-LiF-AlF_3	$3LiAlO_2(c) + AlF_3(c) \rightarrow 3LiF(c) + 2Al_2O_3(c)$	- 26.0
[61]	Li_2O-Al_2O_3-LiF-AlF_3	$LiAlO_2(c) + 3LiF(c) \rightarrow 2Li_2O(c) + AlF_3(c)$	+ 84.8
[62]	Li_2O-Al_2O_3-LiF-AlF_3	$2Li_3AlF_6(\ell) + 3Li_2O(c) \rightarrow 12LiF(c) + Al_2O_3(c)$	-117.5
[63]	Li_2O-Al_2O_3-LiF-AlF_3	$Li_3AlF_6(\ell) + 2Li_2O(c) \rightarrow 6LiF(c) + LiAlO_2(c)$	- 73.4
[64]	Li_2O-Al_2O_3-LiF-AlF_3	$2Li_3AlF_6(\ell) + Al_2O_3(c) \rightarrow 3Li_2O(c) + 4AlF_3(c)$	+162.8
[65]	Li_2O-Al_2O_3-LiF-AlF_3	$Li_3AlF_6(\ell) + 2Al_2O_3(c) \rightarrow 3LiAlO_2(c) + 2AlF_3(c)$	+ 37.3
[66]	LiF-AlF_3-Li-Al	$3Li(\ell) + AlF_3(c) \rightarrow 3LiF(c) + Al(\ell)$	- 73.6
[67]	LiF-AlF_3-Li-Al	$Li_3AlF_6(\ell) + 3LiAl(\ell) \rightarrow 6LiF(c) + 4Al(\ell)$	- 57.8
[68]	LiF-AlF_3-Li-Al	$4Li(\ell) + 2AlF_3(c) \rightarrow Li_3AlF_6(\ell) + LiAl(\ell)$	- 86.5
[69]	LiF-AlF_3-Li-Al	$3LiAl(\ell) + AlF_3(c) \rightarrow 3LiF(c) + 4Al(\ell)$	- 69.1
[70]	LiF-AlF_3-Li-Al	$Li_3AlF_6(\ell) + Al(\ell) \rightarrow 3Li(\ell) + 2AlF_3(c)$	+ 85.0
[71]	LiF-AlF_3-LiAl	$Li_3AlF_6(\ell) + 4Al(\ell) \rightarrow 2AlF_3(c) + 3LiAl(\ell)$	+ 80.4
[72]	LiFAlF_3LiAl	$Li_3AlF_6(\ell) + 3Li(\ell) \rightarrow 6LiF(c) + Al(\ell)$	- 62.3
[73]	LiF-AlF_3-Li-Al	$LiAl(\ell) + 3Li(\ell) \rightarrow 4Li(\ell) + AlF_3(c)$	+ 75.1
[74]	LiF-AlF_3-Li-Al	$LiAl(\ell) + 6LiF(c) \rightarrow 4Li(\ell) + Li_3AlF_6(\ell)$	+ 63.8

Table 4. Calculated Phase Compatibilities in System Li-Al-Mg-O-Cl-F at 1100 K

Reaction #	Subsystem	Reaction	ΔG_R(1100 K), kcal
[75]	$MgO-Li_2O-MgF_2-LiF$	$MgO(c) + 2LiF(\ell) \rightarrow Li_2O(c) + MgF_2(c)$	+ 28.1
[76]	$MgO-Li_2O-MgCl_2-LiCl$	$MgO(c) + 2(MgCl_2 \cdot LiCl)(\ell) \rightarrow 3MgCl_2(\ell) + Li_2O(c)$	+ 52.2
[77]	$MgO-Li_2O-MgCl_2-LiCl$	$MgO(c) + 2LiCl(\ell) \rightarrow MgCl_2(\ell) + Li_2O(c)$	+ 49.1
[78]	$MgO-Li_2O-MgCl_2-LiCl$	$MgO(c) + 3LiCl(\ell) \rightarrow (MgCl_2 \cdot LiCl)(\ell) + Li_2O(c)$	+ 47.6
[79]	$Mg-Li-MgO-Li_2O$	$2Mg(\ell) + Li_2O(c) \rightarrow 2MgLi(\ell) + MgO(c)$	- 9.8
[80]	$Mg-Li-MgO-Li_2O$	$Mg(\ell) + LiO_2(c) \rightarrow MgO(c) + 2Li(\ell)$	- 6.8
[81]	$Mg-Li-MgO-Li_2O$	$MgO(c) + 3Li(\ell) \rightarrow MgLi(\ell) + Li_2O(c)$	+ 5.3
[82]	$MgF_2-LiF-MgCl_2-LiCl$	$3MgCl_2(\ell) + 2LiF(\ell) \rightarrow 2(MgCl_2 \cdot LiCl)(\ell) + MgF_2(c)$	- 24.1
[83]	$MgF_2-LiF-MgCl_2-LiCl$	$3LiCl(\ell) + MgF_2(c) \rightarrow (MgCl_2 \cdot LiCl)(\ell) + 2LiF(c)$	+ 19.6
[84]	$MgF_2-LiF-MgCl_2-LiCl$	$MgCl_2(\ell) + 2LiF(c) \rightarrow 2LiCl(\ell) + MgF_2(c)$	- 21.1
[85]	$Mg-Al-MgO-Al_2O_3$	$3MgO(c) + 2MgAl(\ell) \rightarrow 5Mg(\ell) + Al_2O_3(c)$	+ 30.6
[86]	$Mg-Al-MgO-Al_2O_3$	$4MgO(c) + 2MgAl(\ell) \rightarrow 5Mg(\ell) + MgAl_2O_4(c)$	+ 22.6
[87]	$Mg-Al-MgO-Al_2O_3$	$4MgO(c) + 2Al(\ell) \rightarrow MgAl_2O_4(c) + 3Mg(\ell)$	+ 19.5
[88]	$Mg-Al-MgO-Al_2O_3$	$3MgAl_2O_4(c) + 2Al(\ell) \rightarrow 3Mg(\ell) + 4Al_2O_3(c)$	+ 51.6
[89]	$Mg-Al-MgO-Al_2O_3$	$3MgAl_2O_4(c) + 5Al(\ell) \rightarrow 4Al_2O_3(c) + 3MgAl(\ell)$	+ 47.0
[90]	$Mg-Al-MgO-Al_2O_3$	$3MgO(c) + 3Al(\ell) \rightarrow Al_2O_3(c) + 3MgAl(\ell)$	+ 23.0

Table 4 (continued)

Reaction #	Subsystem	Reaction	ΔG_R(1100 K), kcal
[91]	Mg-Al-MgO-Al_2O_3	$MgAl_2O_4(c) + 3MgAl(\ell) \rightarrow 4MgO(c) + 5Al(\ell)$	- 15.0
[92]	Mg-Al-MgO-Al_2O_3	$3MgAl_2O_4(c) + 2MgAl(\ell) \rightarrow 5Mg(\ell) + 4Al_2O_3(c)$	+ 54.6
[93]	Mg-Al-MgO-Al_2O_3	$3MgO(c) + 2Al(\ell) \rightarrow 3Mg(\ell) + Al_2O_3(c)$	+ 27.5
[94]	Mg-Al-MgF_2-AlF_3	$3MgF_2(c) + 2Al(\ell) \rightarrow 2AlF_3(c) + 3Mg(\ell)$	+ 83.5
[95]	Mg-Al-MgF_2-AlF_3	$3MgF_2(c) + 5Al(\ell) \rightarrow 2AlF_3(c) + 3MgAl(\ell)$	+ 79.0
[96]	Mg-Al-MgF_2-AlF_3	$5Mg(\ell) + 2AlF_3(c) \rightarrow 3MgF_2(c) + 2MgAl(\ell)$	- 86.5
[97]	Mg-Al-$MgCl_2$-$AlCl_3$	$5Mg(\ell) + 2AlCl_3(g) \rightarrow 3MgCl_2(\ell) + 2MgAl(\ell)$	- 88.8
[98]	Mg-Al-$MgCl_2$-$AlCl_3$	$3MgCl_2(\ell) + 2Al(\ell) \rightarrow 3Mg(\ell) + 2AlCl_3(g)$	+ 85.8
[99]	Mg-Al-$MgCl_2$-$AlCl_3$	$3MgCl_2(\ell) + 5Al(\ell) \rightarrow 3MgAl(\ell) + 2AlCl_3(g)$	+ 81.2
[100]	MgF_2-AlF_3-$MgCl_2$-$AlCl_3$	$MgCl_2(\ell) + AlF_3(c) \rightarrow MgF_2(c) + AlCl_3(g)$	+ 56.3
[101]	Mg-Li-MgF_2-LiF	$3Mg(\ell) + 2LiF(c) \rightarrow MgF_2(c) + 2MgLi(\ell)$	+ 18.2
[102]	Mg-Li-MgF_2-LiF	$MgLi(\ell) + 2LiF(c) \rightarrow MgF_2(c) + 3Li(\ell)$	+ 22.8
[103]	Mg-Li-MgF_2-LiF	$MgF_2(c) + 2Li(\ell) \rightarrow 2LiF(c) + Mg(\ell)$	- 21.2
[104]	MgO-Al_2O_3-$MgCl_2$-$AlCl_3$	$4MgO(c) + AlCl_3(g) \rightarrow MgCl_2(\ell) + MgAl_2O_4(c)$	+ 33.0
[105]	MgO-Al_2O_3-$MgCl_2$-$AlCl_3$	$3MgCl_2(\ell) + Al_2O_3(c) \rightarrow 3MgO(c) + 2AlCl_3(g)$	+ 58.2
[106]	MgO-Al_2O_3-$MgCl_2$-$AlCl_3$	$3MgAl_2O_4(c) + 2AlCl_3(g) \rightarrow 3MgCl_2(\ell) + 4Al_2O_3(c)$	- 34.2

Table 4 (continued)

Reaction #	Subsystem	Reaction	ΔG_R(1100 K), kcal
[107]	$MgO-Al_2O_3-MgF_2-AlF_3$	$3MgF_2(c) + MgAl_2O_4(c) \rightarrow 4MgO(c) + 2AlF_3(c)$	+ 64.0
[108]	$MgO-Al_2O_3-MgF_2-AlF_3$	$3MgO(c) + 2AlF_3(c) \rightarrow 3MgF_2(c) + Al_2O_3(c)$	- 56.0
[109]	$MgO-Al_2O_3-MgF_2-AlF_3$	$2AlF_3(c) + 3MgAl_2O_4(c) \rightarrow 3MgF_2(c) + 4Al_2O_3(c)$	- 31.9
[110]	$Mg-Li-MgCl_2-LiCl$	$3MgCl_2(\ell) + 2Li(\ell) \rightarrow 2(MgCl_2 \cdot LiCl)(\ell) + Mg(\ell)$	- 45.4
[111]	$Mg-Li-MgCl_2-LiCl$	$MgCl_2(\ell) + 2Li(\ell) \rightarrow Mg(\ell) + 2LiCl(\ell)$	- 42.3
[112]	$Mg-Li-MgCl_2-LiCl$	$(MgCl_2 \cdot LiCl)(\ell) + 2Li(\ell) \rightarrow 3LiCl(\ell) + Mg(\ell)$	- 40.8
[113]	$MgO-Al_2O_3-Li_2O-MgCl_2-AlCl_3-LiCl$	$MgCl_2(\ell) + 2LiAlO_2(c) \rightarrow MgAl_2O_4(c) + 2LiCl(\ell)$	- 27.8
[114]	$MgO-Al_2O_3-Li_2O-MgF_2-AlF_3-LiF$	$MgF_2(c) + 2LiAlO_2(c) \rightarrow MgAl_2O_4(c) + 2LiF(c)$	- 6.7
[115]	$MgO-Al_2O_3-Li_2O-MgF_2-AlF_3-LiF$	$Li_3AlF_6(\ell) + 3MgAl_2O_4(c) \rightarrow 3LiAlO_2(c) + 3MgF_2(c) + 2Al_2O_3(c)$	+ 5.4
[116]	$MgO-Al_2O_3-Li_2O-MgF_2-AlF_3-LiF$	$2Li_3AlF_6(\ell) + 6MgAl_2O_4(c) \rightarrow 6MgF_2(c) + 6LiAlO_2(c) + 4Al_2O_3(c)$	+ 10.8
[117]	$MgO-Al_2O_3-Li_2O-MgF_2-AlF_3-LiF$	$Li_3AlF_6(\ell) + 2MgO \rightarrow 2LiF(c) + 2MgF_2(c) + LiAlO_2(c)$	- 17.3

Table 4 (continued)

Reaction #	Subsystem	Reaction	ΔG_R(1100 K), kcal
[118]	MgF_2-AlF_3-LiF-$MgCl_2$-$AlCl_3$-LiCl	$3MgCl_2(\ell) + 2Li_3AlF_6(\ell) \rightarrow 6LiCl(\ell) + 3MgF_2(c) + 2AlF_3(c)$	- 40.6
[119]	Mg-Al-Li-MgF_2-AlF_3-LiF	$2Li_3AlF_6(\ell) + 3Mg(\ell) \rightarrow 6LiF(c) + 3MgF_2(c) + 2Al(\ell)$	- 60.9
[120]	Mg-Al-Li-MgO-Al_2O_3-Li_2O	$LiAlO_2(c) + 2Mg(\ell) \rightarrow 2MgO(c) + LiAl(\ell)$	- 4.0
[121]	Mg-Al-Li-MgO-Al_2O_3-Li_2O	$MgAl_2O_4(c) + 2Li(\ell) \rightarrow 2LiAlO_2(c) + Mg(\ell)$	- 14.6
[122]	Mg-Al-Li-MgO-Al_2O_3-Li_2O	$2MgAl_2O_4(c) + 3Li(\ell) \rightarrow 2MgO(c) + 3LiAlO_2(c) + Al(\ell)$	- 31.6

Table 5. Progressive Oxidation of LiAlMg alloy by reaction with Components of ClO_3F

Reaction #	Type of Reaction	Reaction
[123]	chlorination	$LiAlMg + ClO_3F \rightarrow Mg + Al + LiCl + (1/2)F_2 + (3/2)O_2$
[124]	chlorination	$LiAlMg + 3ClO_3F \rightarrow MgCl_2 + LiCl + Al + (3/2)F_2 + (9/2)O_2$
[125]	chlorination	$LiAlMg + 6ClO_3F \rightarrow LiCl + MgCl_2 + AlCl_3(g) + 3F_2 + 9O_2$
[126]	fluorination	$LiAlMg + ClO_3F \rightarrow Mg + Al + LiF + (1/2)F_2 + (3/2)O_2$
[127]	fluorination	$3LiAlMg + 12ClO_3F \rightarrow 3MgF_2 + Li_3AlF_6 + 2Al + 6Cl_2 + 18O_2$
[128]	fluorination	$3LiAlMg + 18ClO_3F \rightarrow 3MgF_2 + 2AlF_3 + Li_3AlF_6 + 9Cl_2 + 27O_2$
[129]	oxygenation	$LiAlMg + (2/3)ClO_3F \rightarrow Mg + LiAlO_2 + (2/3)Cl_2 + (1/3)F_2$
[130]	oxygenation	$LiAlMg + ClO_3F \rightarrow MgO + LiAlO_2 + (1/2)Cl_2 + (1/2)F_2$
[131]	chlorination & oxygenation	$7LiAlMg + 6ClO_3F \rightarrow 6LiCl + 3MgAl_2O_4 + LiAlO_2 + 4MgO + 3F_2$
[132]	fluorination & oxygenation	$7LiAlMg + 6ClO_3F \rightarrow 6LiF + 3MgAl_2O_4 + LiAlO_2 + 4MgO + 3Cl_2$
[133]	chlorination & fluorination	$LiAlMg + 3ClO_3F \rightarrow LiCl + AlF_3 + MgCl_2 + (9/2)O_2$
[134]	chlorination & fluorination & oxygenation	$4LiAlMg + 3ClO_3F \rightarrow MgF_2 + LiF + 3LiCl + 2MgAl_2O_4 + MgO$

Table 6. Calculated Phase Compatibilities in System Li-Be-O-Cl-F at 1100 K

Reaction #	Subsystem	Reaction	ΔG_R(1100 K), kcal
[135]	Li-Be-LiF-BeF_2	$2Li(\ell) + Li_2BeF_4(\ell) \rightarrow 4LiF(c) + Be(c)$	- 28.3
[136]	Li-Be-LiF-BeF_2	$2Li(\ell) + 4LiBeF_3(\ell) \rightarrow 3Li_2BeF_4(\ell) + Be(c)$	- 42.1
[137]	Li-Be-LiF-BeF_2	$2Li(\ell) + LiBeF_3(\ell) \rightarrow 3LiF(c) + Be(c)$	- 31.7
[138]	Li-Be-LiF-BeF_2	$2Li(\ell) + BeF_2(\ell) \rightarrow 2LiF(c) + Be(c)$	- 38.6
[139]	Li-Be-LiF-BeF_2	$2Li(\ell) + 2BeF_2(\ell) \rightarrow Li_2BeF_4(\ell) + Be(c)$	- 48.9
[140]	Li-Be-LiF-BeF_2	$2Li(\ell) + 3BeF_2(\ell) \rightarrow 2LiBeF_3(\ell) + Be(c)$	- 52.2
[141]	Li_2O-BeO-LiF-BeF_2	$Li_2O(c) + Li_2BeF_4(\ell) \rightarrow 4LiF(c) + BeO(c)$	- 39.7
[142]	Li_2O-BeO-LiF-BeF_4	$Li_2O(c) + 4LiBeF_3(\ell) \rightarrow 3Li_2BeF_4(\ell) + BeO(c)$	- 53.5
[143]	Li_2O-BeO-LiF-BeF_4	$Li_2O(c) + LiBeF_3(\ell) \rightarrow 3LiF(c) + BeO(c)$	- 43.1
[144]	Li_2O-BeO-LiF-BeF_4	$Li_2O(c) + BeF_2(\ell) \rightarrow 2LiF(c) + BeO(c)$	- 50.0
[145]	Li_2O-BeO-LiF-BeF_4	$Li_2O(c) + 2BeF_2(\ell) \rightarrow BeO(c) + Li_2BeF_4(\ell)$	- 60.3
[146]	Li_2O-BeO-LiF-BeF_4	$Li_2O(c) + 3BeF_2(\ell) \rightarrow BeO(c) + 2LiBeF_3(\ell)$	- 63.6
[147]	Li-Be-LiCl-$BeCl_2$	$2LiCl(\ell) + Be(c) \rightarrow BeCl_2(g) + 2Li(\ell)$	+ 63.9
[148]	Li-Be-Li_2O-BeO	$2Li(\ell) + BeO(c) \rightarrow Li_2O(c) + Be(c)$	+ 11.4
[149]	Li_2O-BeO-LiCl-$BeCl_2$	$Li_2O(c) + BeCl_2(g) \rightarrow 2LiCl(\ell) + BeO(c)$	- 75.3
[150]	LiCl-$BeCl_2$-LiF-BeF_2	$2LiCl(\ell) + Li_2BeF_4(\ell) \rightarrow 4LiF(c) + BeCl_2(g)$	+ 35.6

Table 6 (continued)

Reaction #	Subsystem	Reaction	ΔG_R(1100 K), kcal
[151]	$LiCl-BeCl_2-LiF-BeF_2$	$2LiCl(\ell) + LiBeF_3(\ell) \rightarrow 3LiF(c) + BeCl_2(g)$	+ 32.1
[152]	$LiCl-BeCl_2-LiF-BeF_2$	$2LiCl(\ell) + BeF_2(\ell) \rightarrow 2LiF(c) + BeCl_2(g)$	+ 25.3
[153]	$LiCl-BeCl_2-LiF-BeF_2$	$Li_2BeF_4(\ell) + BeCl_2(g) \rightarrow 2LiCl(\ell) + 2BeF_2(\ell)$	- 15.0
[154]	$LiCl-BeCl_2-LiF-BeF_2$	$2LiCl(\ell) + 4LiBeF_3(\ell) \rightarrow 3Li_2BeF_4(\ell) + BeCl_2(g)$	+ 21.8
[155]	$LiCl-BeCl_2-LiF-BeF_2$	$2LiBeF_3(\ell) + BeCl_2(g) \rightarrow 2LiCl(\ell) + 3BeF_2(\ell)$	- 11.6

Table 7. Progressive Oxidation of (Li+Be) Fuel by Reaction with Components of ClO_3F

Reaction #	Type of Reaction	Reaction
[156]	fluorination	$Li + Be + ClO_3F \rightarrow LiF + Be + (1/2)Cl_2 + (3/2)O_2$
[157]	fluorination	$Li + Be + 2ClO_3F \rightarrow LiF + BeF_2 + Cl_2 + 3O_2$
[158]	chlorination	$Li + Be + ClO_3F \rightarrow LiCl + Be + (1/2)F_2 + (3/2)O_2$
[159]	chlorination	$Li + Be + 3ClO_3F \rightarrow LiCl + BeCl_2(g) + (3/2)F_2 + (9/2)O_2$
[160]	oxygenation	$3Li + 3Be + ClO_3F \rightarrow 3Li + 3BeO + (1/2)Cl_2 + (1/2)F_2$
[161]	oxygenation	$3Li + 3Be + ClO_3F \rightarrow (3/2)Li_2O + 3BeO + (1/2)Cl_2 + (1/2)F_2$
[162]	fluorination & oxygenation	$3Li + 3Be + ClO_3F \rightarrow LiF + 2Li + 3BeO + (1/2)Cl_2$
[163]	fluorination & oxygenation	$3Li + 3Be + ClO_3F \rightarrow 2Li_2O + 3BeO + (1/2)Cl_2 + (1/2)F_2$
[164]	chlorination & oxygenation	$3Li + 3Be + ClO_3F \rightarrow 2Li + 3BeO + LiCl + (1/2)F_2$
[165]	chlorination & oxygenation	$7Li + 7Be + 3ClO_3F \rightarrow 2Li_2O + 3LiCl + 7BeO + (3/2)F_2$
[166]	chlorination & fluorination	$2Li + 2Be + ClO_3F \rightarrow LiF + LiCl + 2Be + (3/2)O_2$
[167]	chlorination & fluorination	$2Li + 2Be + 2ClO_3F \rightarrow Be + BeF_2 + 2LiCl + 3O_2$
[168]	chlorination & fluorination	$4Li + 4Be + 6ClO_3F \rightarrow 3BeF_2 + 4LiCl + BeCl_2(g) + 9O_2$
[169]	chlorination & fluorination & oxygenation	$8Li + 8Be + 3ClO_3F \rightarrow 8BeO + Li_2O + 3LiF + 3LiCl$

Table 8. Calculated Phase Compatibilities in System Li-C-F at 1100 K

Reaction #	Subsystem	Reaction	ΔG_R(1100 K), kcal
[170]	Li-C-F	$10Li(\ell) + 2CF_4(g) \rightarrow Li_2C_2(c) + 8LiF(c)$	-620.7
[171]	Li-C-F	$4LiF(c) + 5C(c) \rightarrow 2Li_2C_2(c) + CF_4(g)$	+284.5
[172]	Li-C-F	$Li_2C_2(c) + 5F_2(g) \rightarrow 2CF_4(g) + 2LiF(c)$	-600.0
[173]	Li-C-F	$2C(c) + 2LiF(c) \rightarrow Li_2C_2(c) + 2F(g)$	+233.8
[174]	Li-C-F	$Li_2C_2(c) + 8F(g) \rightarrow 2Li(\ell) + 2CF_4(g)$	-355.9
[175]	Li-C-F	$4LiF(c) + C(g) \rightarrow 4Li(\ell) + CF_4(g)$	+305.2

Table 9. Calculated Phase Compatibilities in System Li-B-N-F at 1100 K

Reaction #	Subsystem	Reaction	ΔG_R (1100 K), kcal
[177]	Li-B-LiF-BF_3	$4Li(\ell) + BF_3(g) \rightarrow 3LiF(c) + LiB(c)$	-112.0
[178]	Li-B-LiF-BF_3	$3LiF(c) + B(c) \rightarrow 3Li(\ell) + BF_3(g)$	+110.5
[179]	Li-B-LiF-BF_3	$3LiF(c) + 3B(c) \rightarrow 3LiB(c) + BF_3(g)$	+106.0
[180]	Li-B-Li_3N-BN	$Li_3N(c) + LiB(c) \rightarrow 4Li(\ell) + BN(c)$	- 31.0
[181]	Li-B-Li_3N-BN	$Li_3N(c) + B(c) \rightarrow 3Li(\ell) + BN(c)$	- 32.5
[182]	Li-B-Li_3N-BN	$3LiB(c) + BN(c) \rightarrow Li_3N(c) + 4B(c)$	+ 37.1
[183]	Li_3N-BN-LiF-BF_3	$3LiF(c) + BN(c) \rightarrow Li_3N(c) + BF_3(g)$	+143.0

Table 10. Progressive Oxidation of LiB by NF_3

Reaction #	Reaction
[184]	$3LiB + NF_3 \rightarrow 2B + 3LiF + BN$
[185]	$3LiB + 2NF_3 \rightarrow 3LiF + BF_3 + 2BN$

Table 11. Comparison of Selected Liquid Metal Stored Chemical Energy Reactions

Reaction #	Primary Fuel	1st Stage Oxidant	1st Stage Reaction Products	2nd Stage Oxidant	2nd Stage Reaction Products	Total Calculated Enthalpy Yield at 1100 K in kcal/g (fuel & oxidant)*
[14]	Li	H_2O	Li_2O, H_2	O_2	H_2O	4.8
[169]	Li + Be	ClO_3F	BeO, Li_2O, LiF, LiCl	-	-	4.6
[186]	2Li + LiB	NF_3	LiF, BN	-	-	4.6
[2]	LiAl	H_2O	$LiAlO_2$, H_2	O_2	H_2O	4.3
[188]	Al	H_2O	Al_2O_3, H_2	O_2	H_2O	4.0
[21]	MgAl	H_2O	MgO, $MgAl_2O_4$, H_2	O_2	H_2O	3.8
[74x]	LiAl	ClO_3F	Al_2O_3, LiF, LiCl	-	-	3.8
[187]	Mg	H_2O	MgO, H_2	O_2	H_2O	3.6
[134]	LiAlMg	ClO_3F	MgF_2, LiF, LiCl, $MgAl_2O_4$, MgO	-	-	3.6
[13]	Li	H_2O	LiOH, H_2	O_2	H_2O	3.5
[176]	Li	$C_{11}F_{20}$	C, LiF	-	-	3.1**
[1]	Li	SF_6	LiF, Li_2S	-	-	3.0***
[20]	Li	H_2O	LiOH, H_2	$2NaO_2 + H_2O \rightarrow 2NaOH(3/2)O_2$	H_2O	2.4

* Calculation ignores weight of recycled water in dual stage reactions

** Calculated at 298 K

*** From ref. (11)

XV. Figures

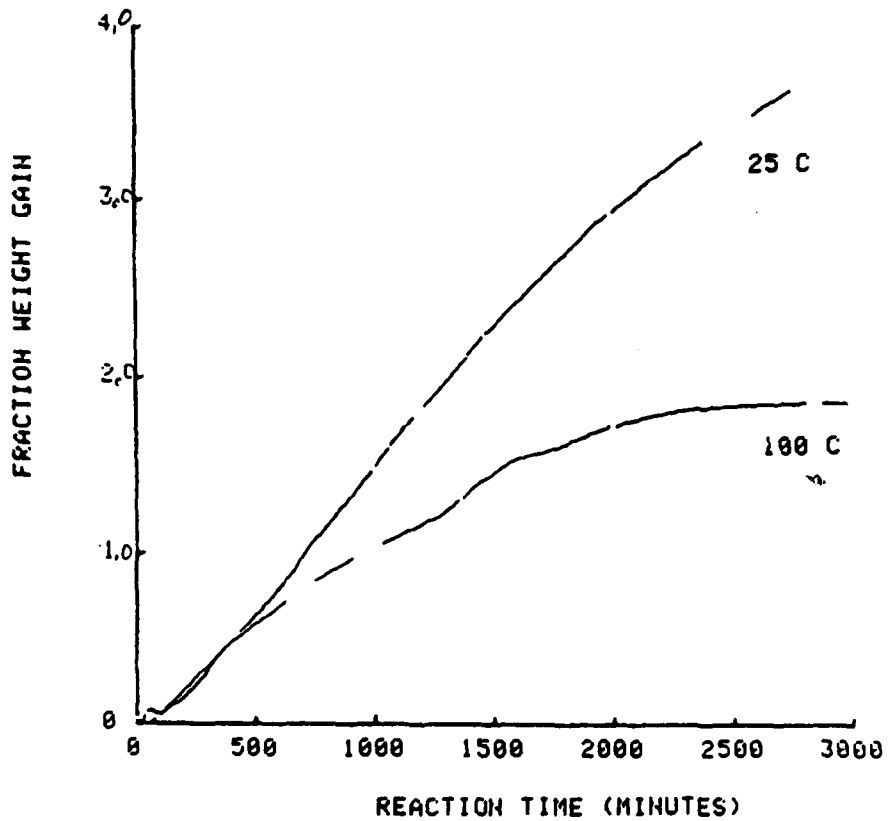

Figure 1. Thermogravimetric analysis of reaction of powdered lithium metal with flowing water-saturated argon at 25 and 100 °C.

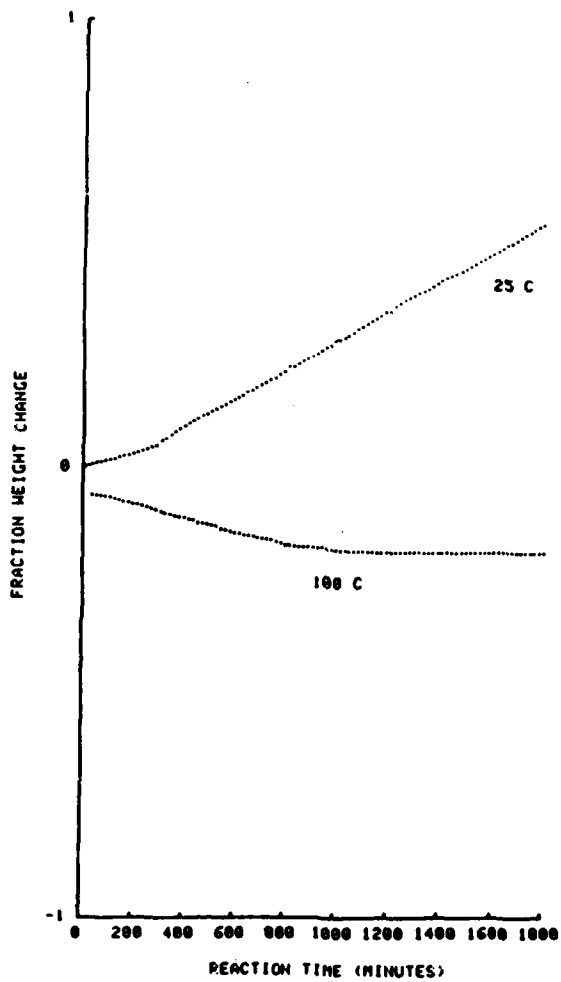

Figure 2. Thermogravimetric analysis of reaction of powdered sodium superoxide with flowing water-saturated argon at 25 and 100 °C.

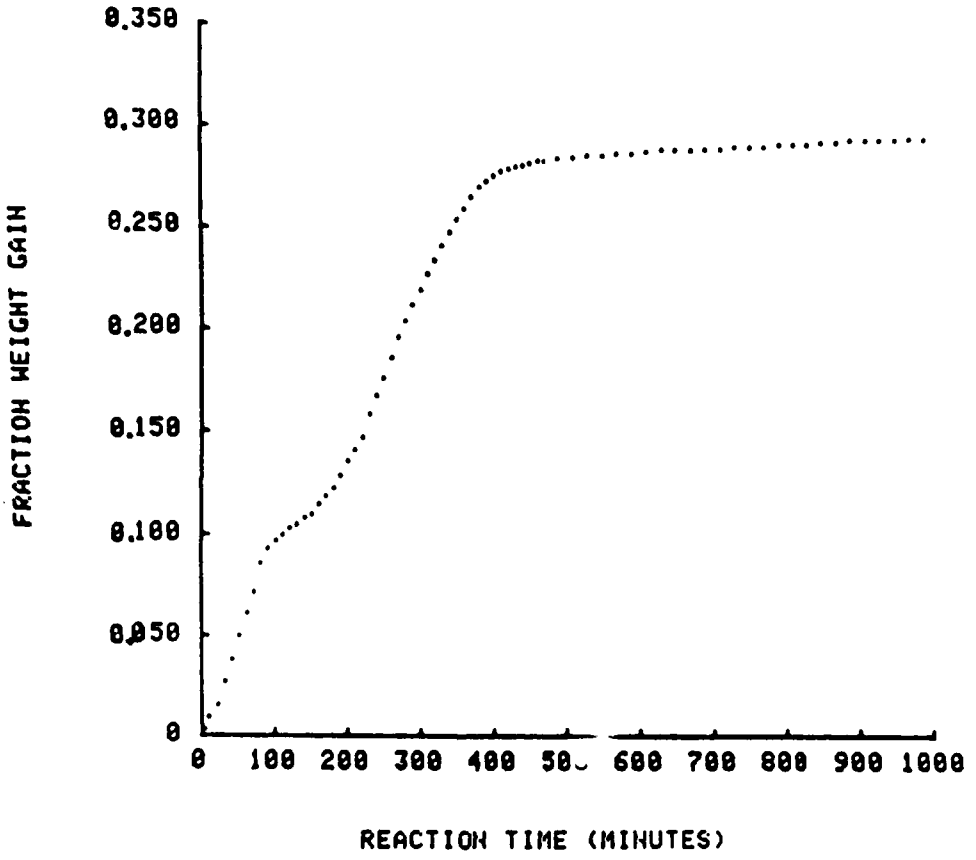

Figure 3. Thermogravimetric analysis of reaction of powdered MgAl alloy with flowing water-saturated argon at 650 °C.

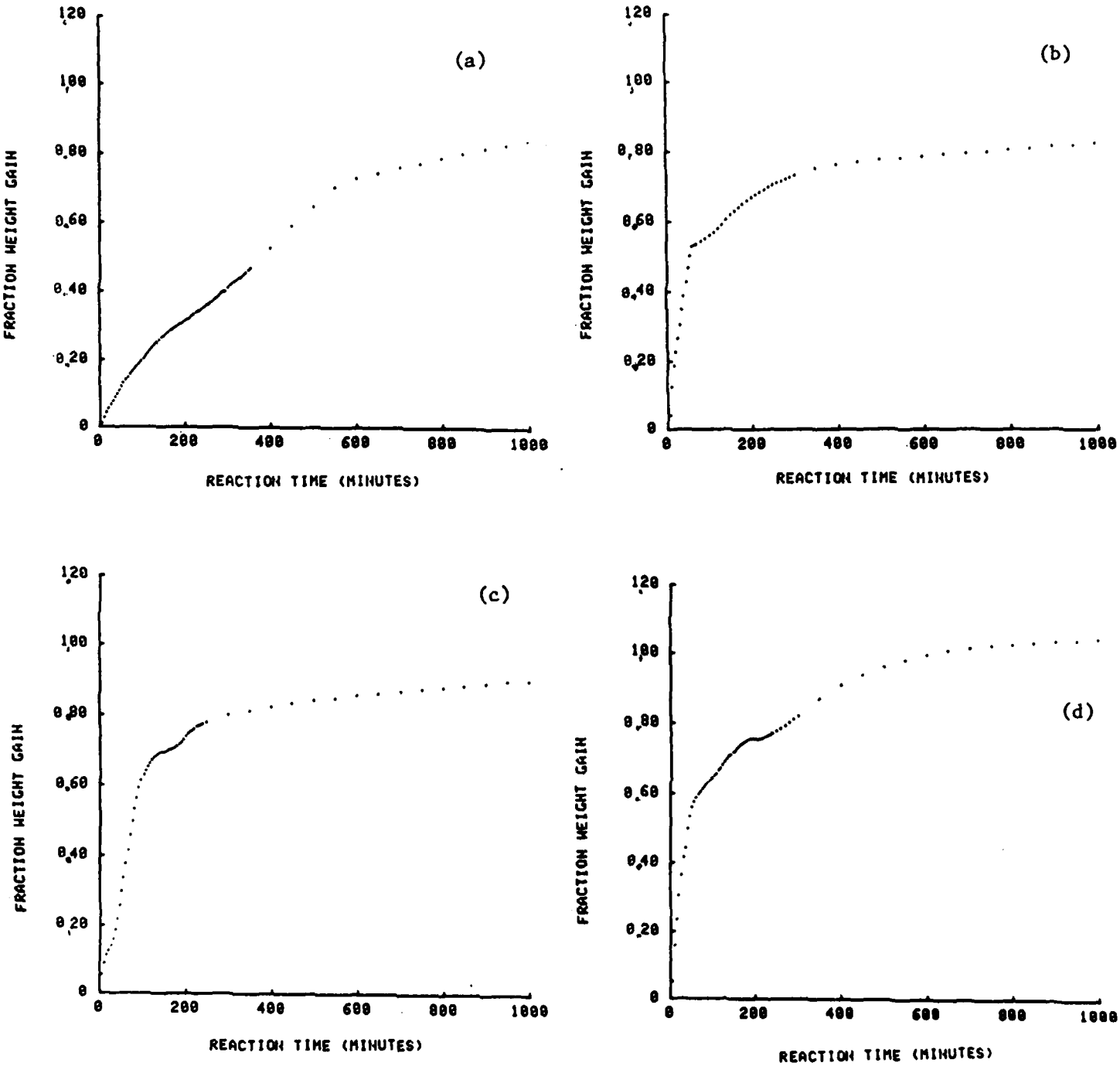

Figure 4. Thermogravimetric analysis of reaction of powdered LiAl alloy with ClO_3F. (a) 350 °C; (b) 450 °C; (c) 550 °C; (d) 650 °C.

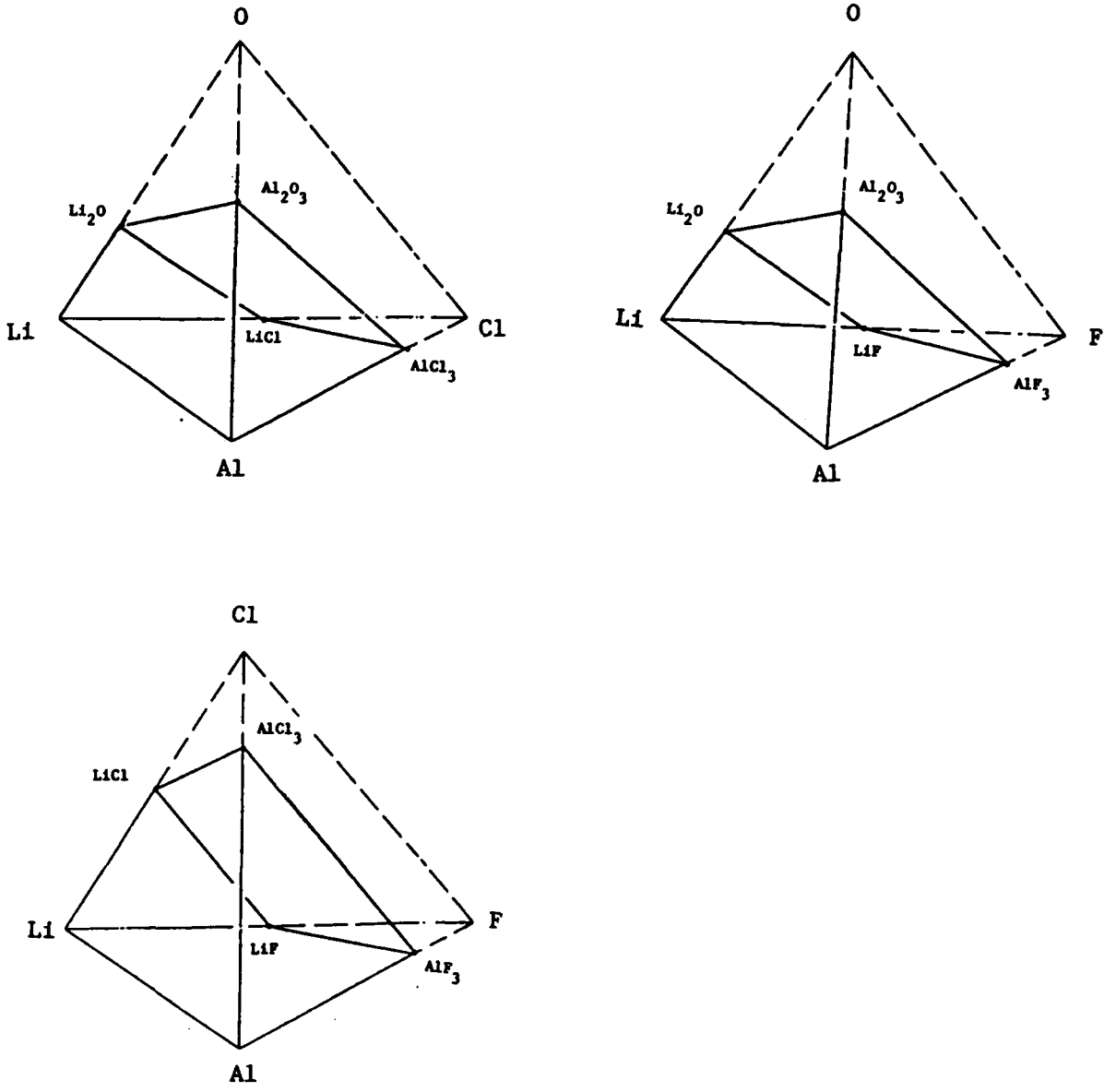

Figure 5. Quaternary reciprocal systems relevant to oxidation of LiAl alloy by ClO_3F.

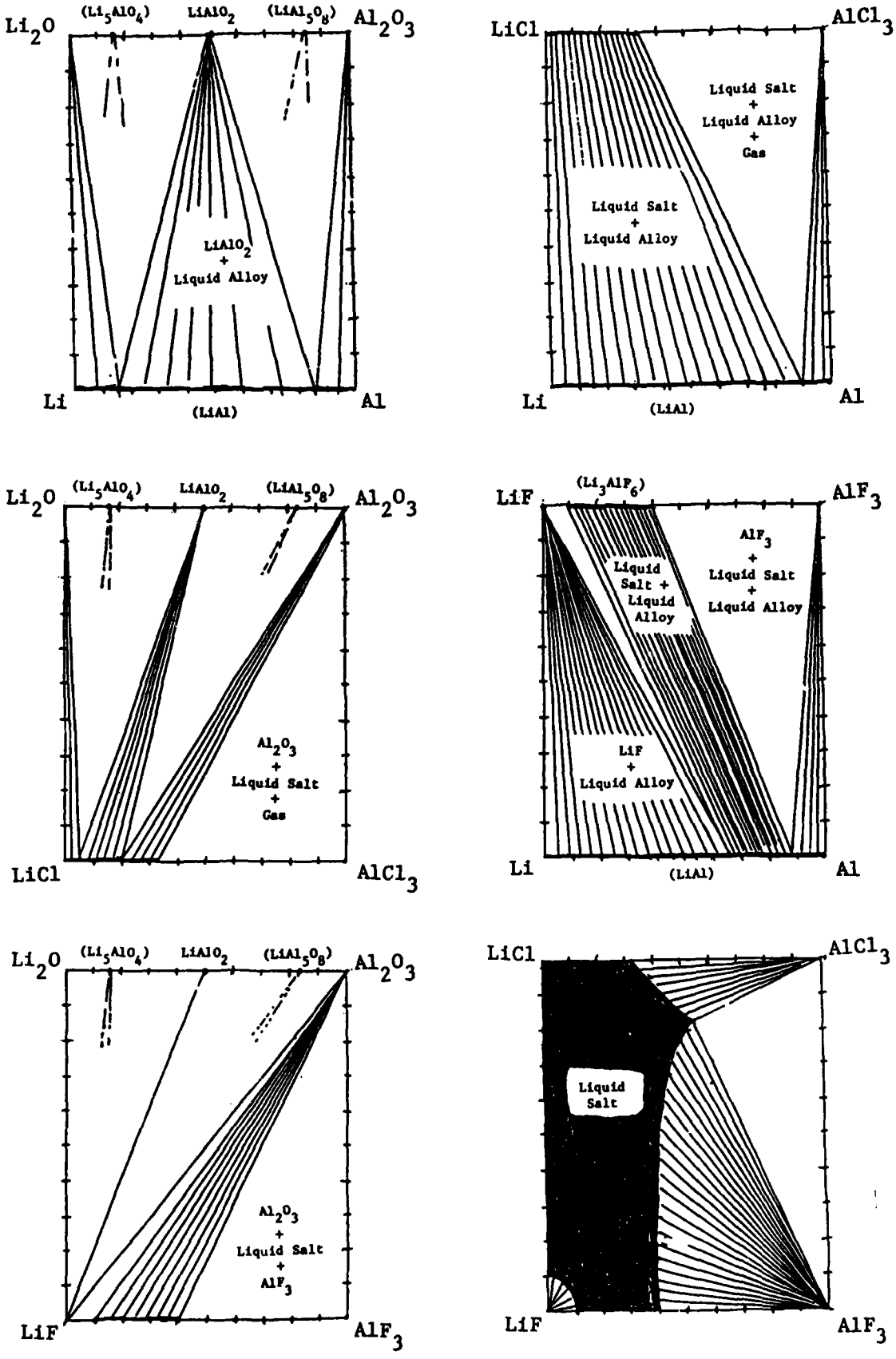

Figure 6. Calculated phase compatibilities in ternary reciprocal subsystems of the system Li-Al-Cl-O-F at 1100 K, 1 atm.

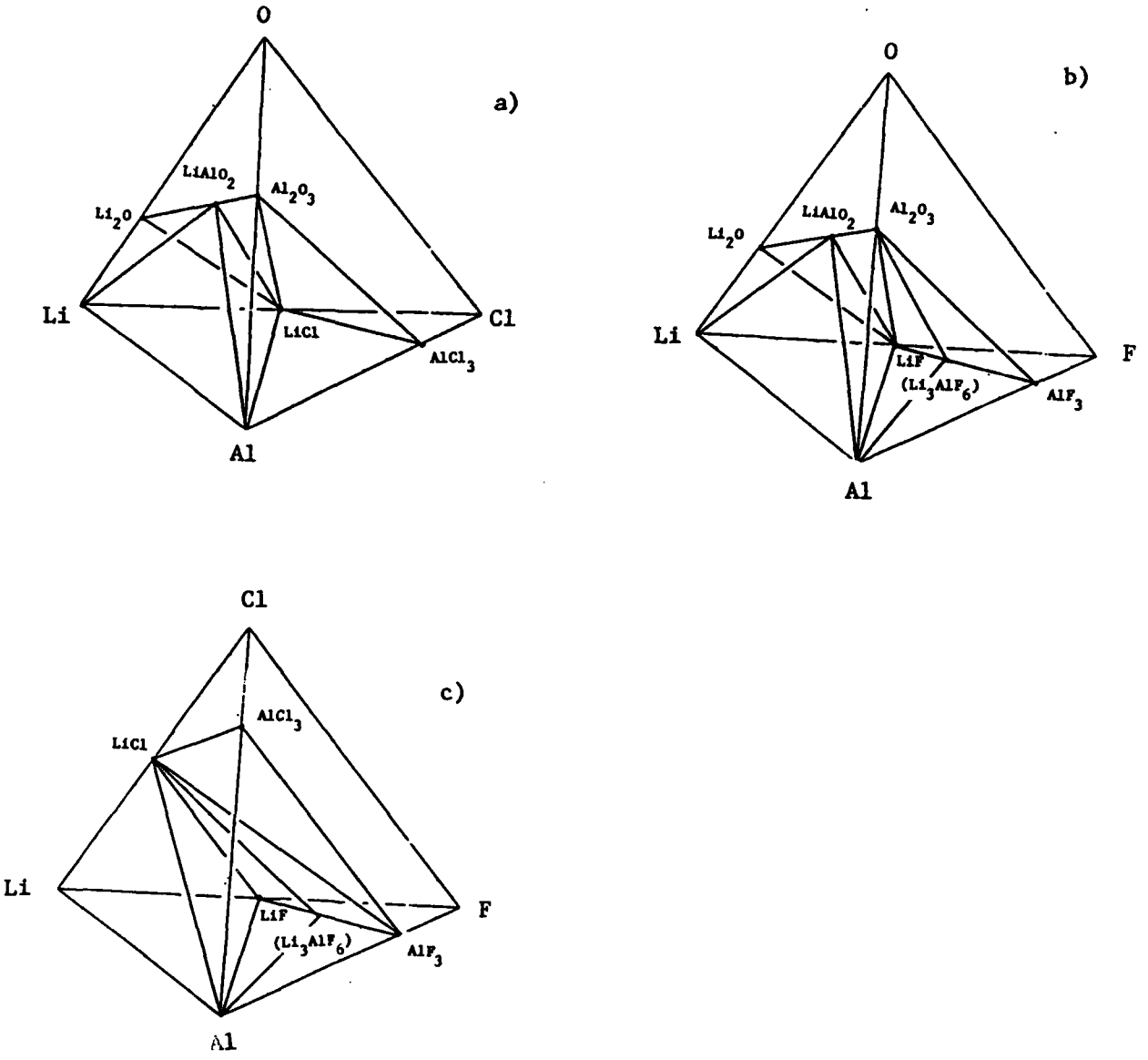

Figure 7. Outline of 1100 K, 1 atm phase compatibilities relating to oxidation of LiAl alloy by ClO_3F. (a) chlorination and oxygenation; (b) fluorination and oxygenation; (c) fluorination and chlorination.

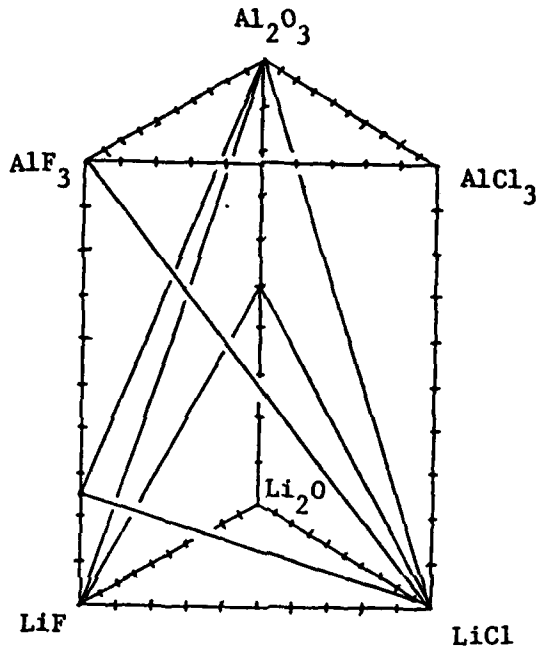

Figure 8. Outline of 1100 K, 1 atm phase compatibilities among products of complete oxidation of LiAl alloy by ClO_3F.

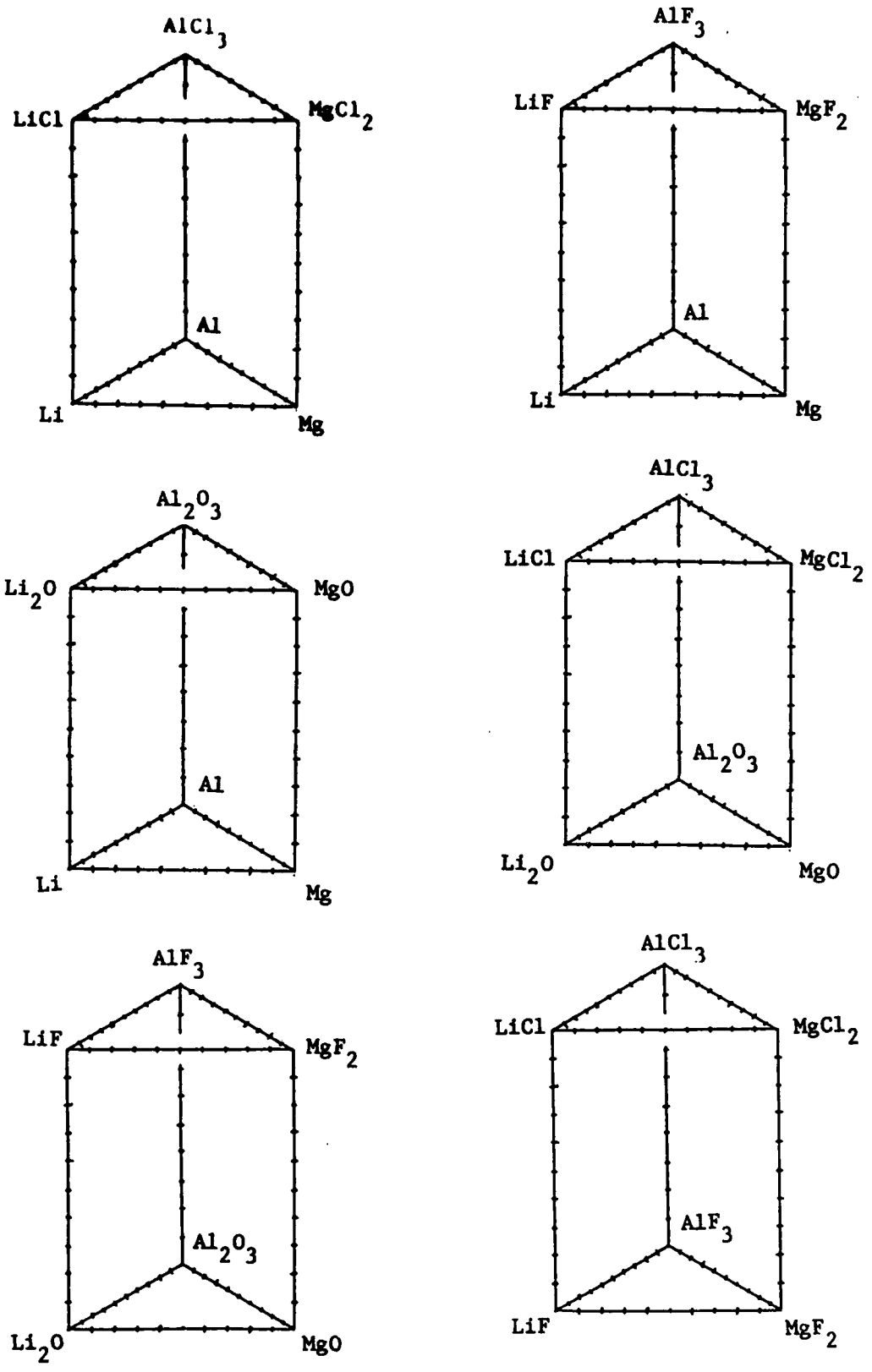

Figure 9. Quaternary reciprocal systems relevant to oxidation of LiAlMg alloy by ClO_3F.

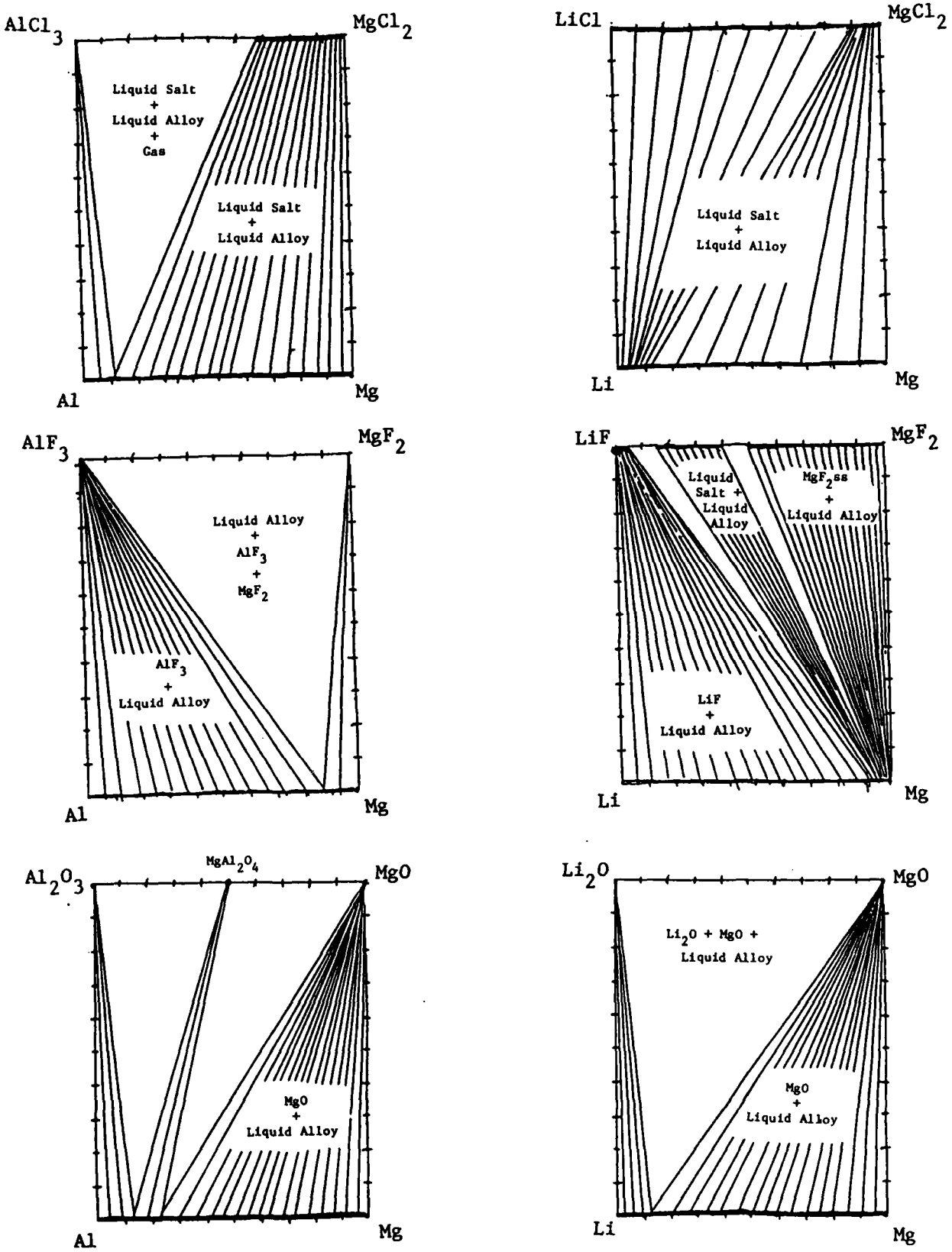

Figure 10. Calculated phase compatibilities in ternary reciprocal subsystems of the system Li-Al-Mg-Cl-O-F at 1100 K, 1 atm.

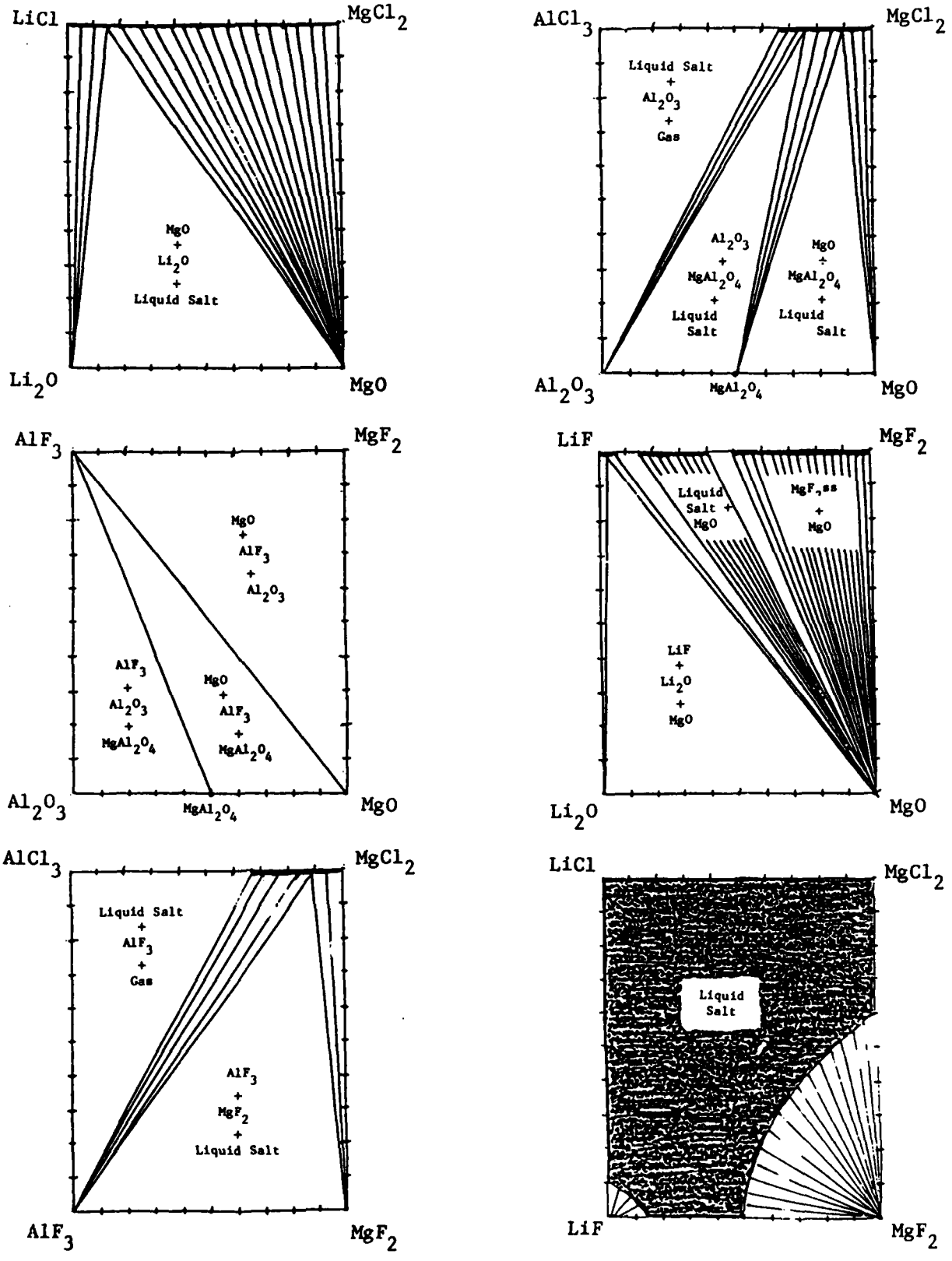

Figure 10, continued.

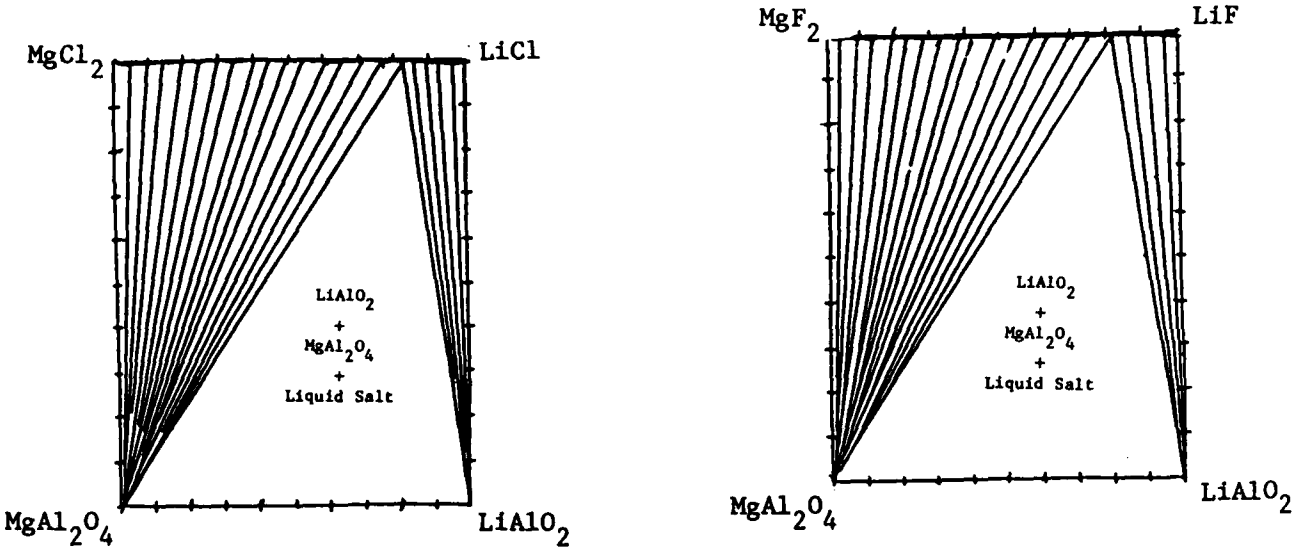

Figure 10, continued.

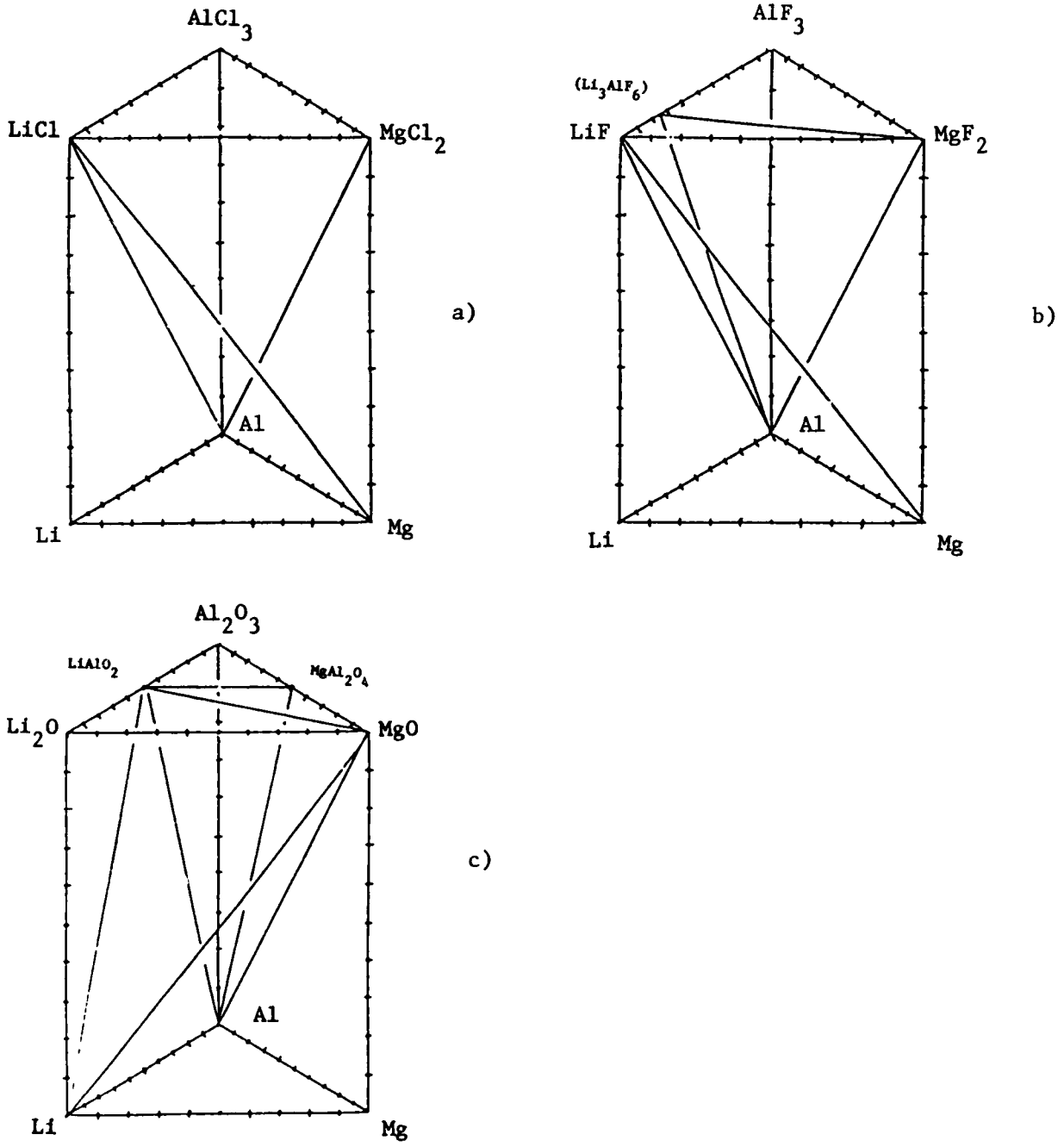

Figure 11. Outline of 1100 K, 1 atm phase compatibilities relating to oxidation of LiAlMg alloy by ClO_3F. (a) chlorination; (b) fluorination; (c) oxygenation.

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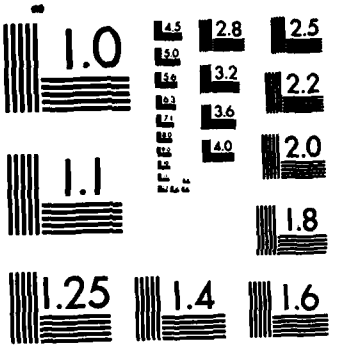

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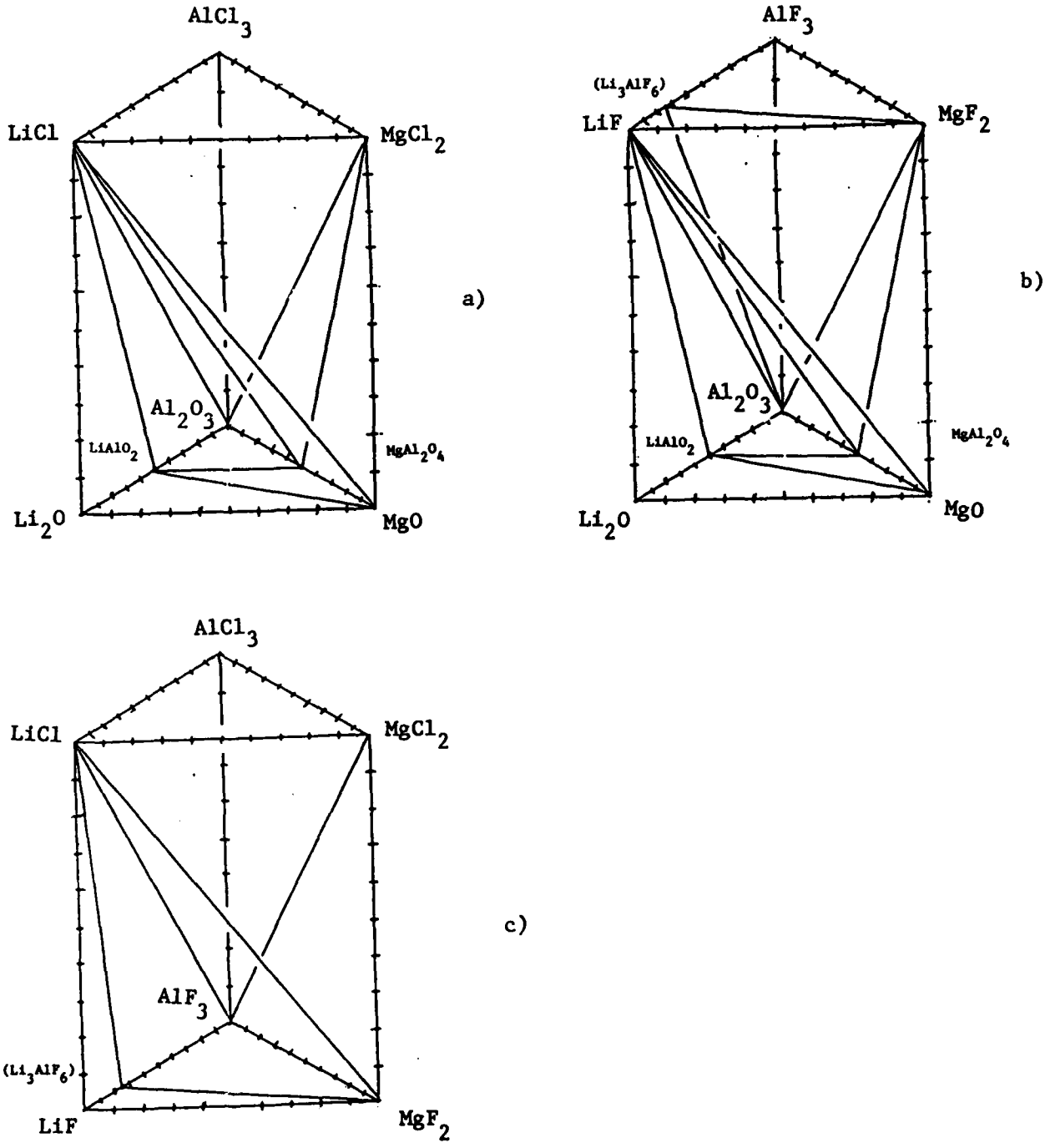

Figure 12. Outline of 1100 K, 1 atm phase compatibilities among products of complete oxidation of LiAlMg alloy by ClO_3F. (a) chlorination and oxygenation; (b) fluorination and oxygenation; (c) fluorination and chlorination

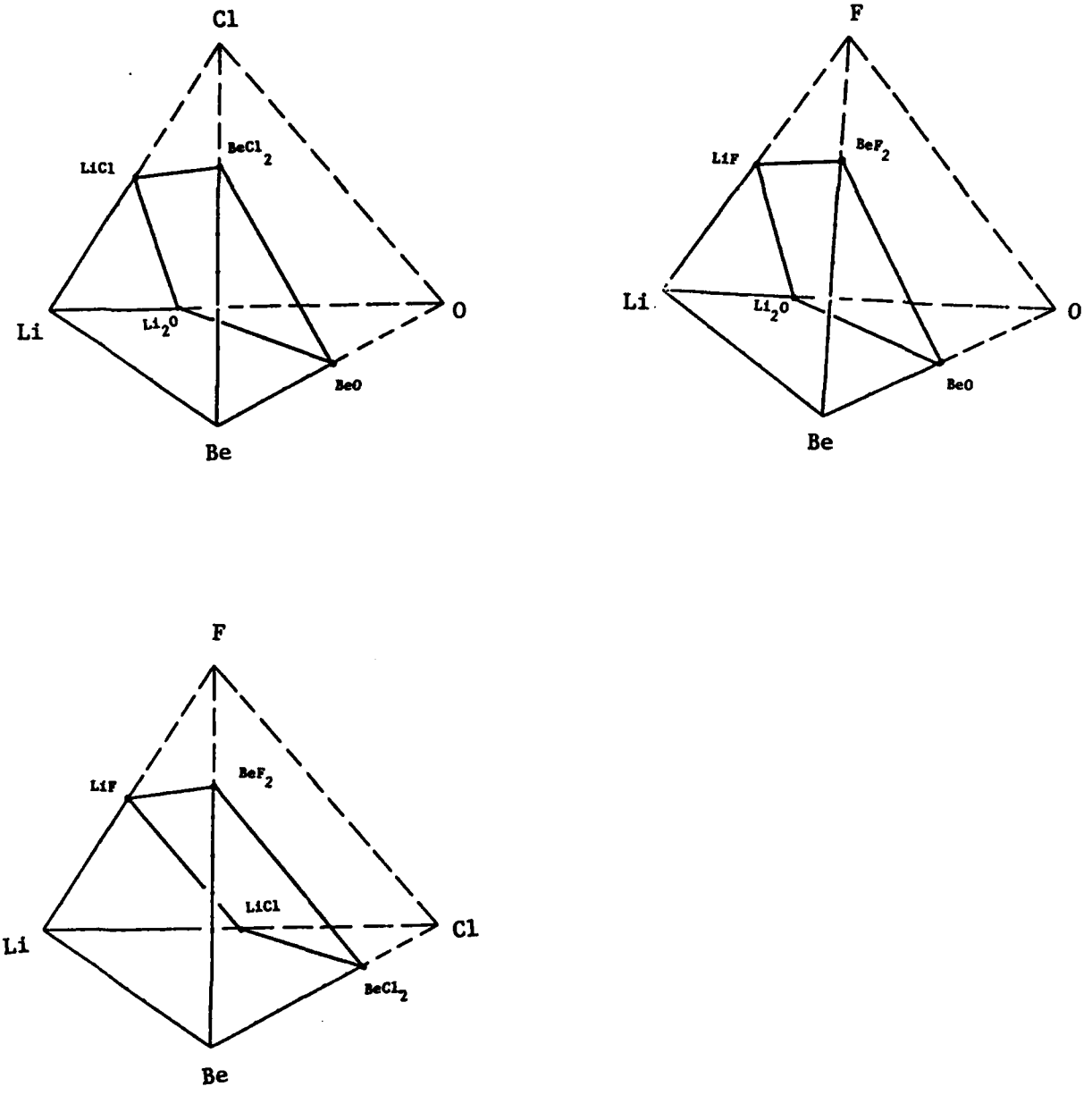

Figure 13. Quaternary reciprocal systems relevant to oxidation of Li/Be mixtures by ClO_3F.

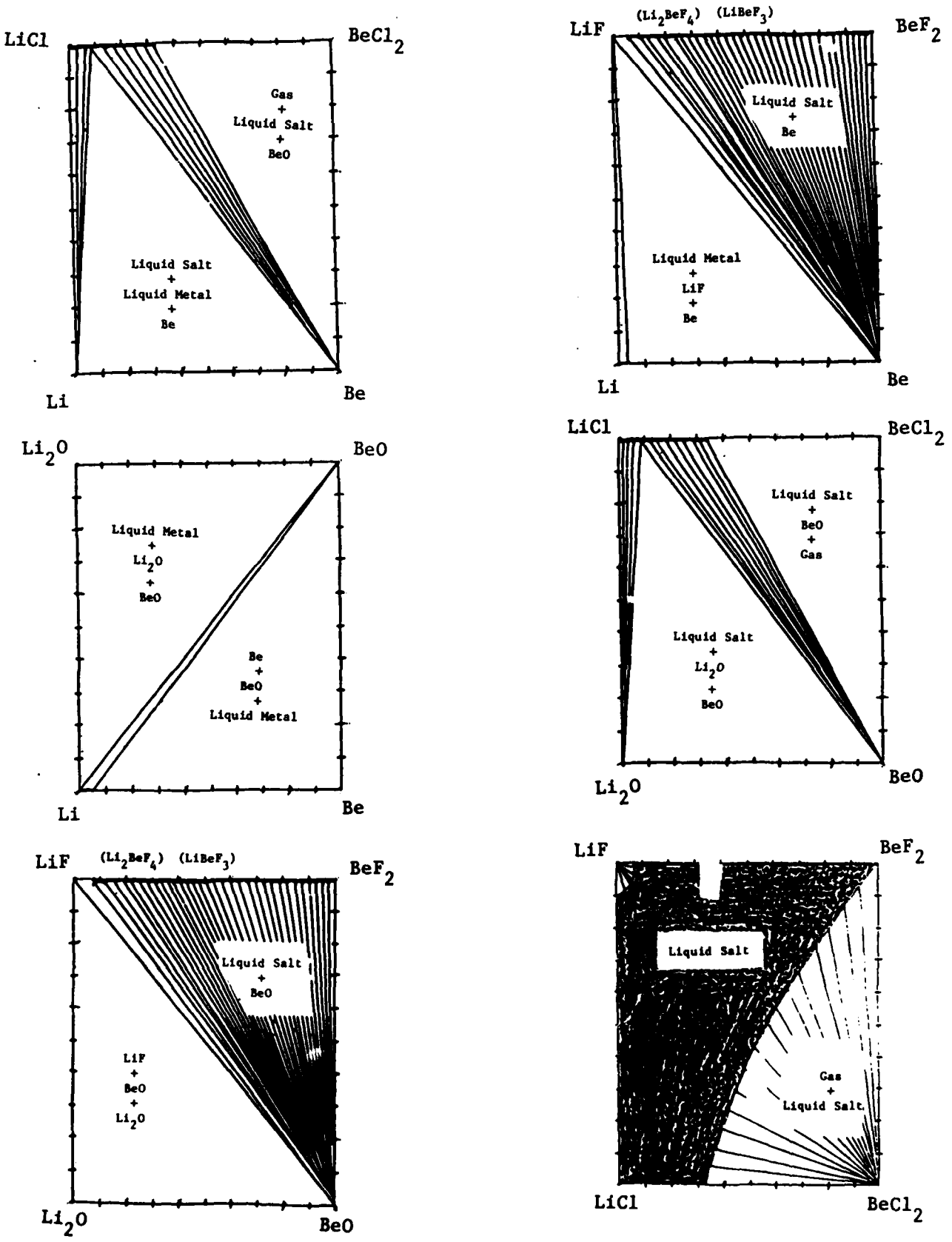

Figure 14. Calculated phase compatibilities in ternary reciprocal subsystems of the system Li-Be-Cl-O-F at 1100 K, 1 atm.

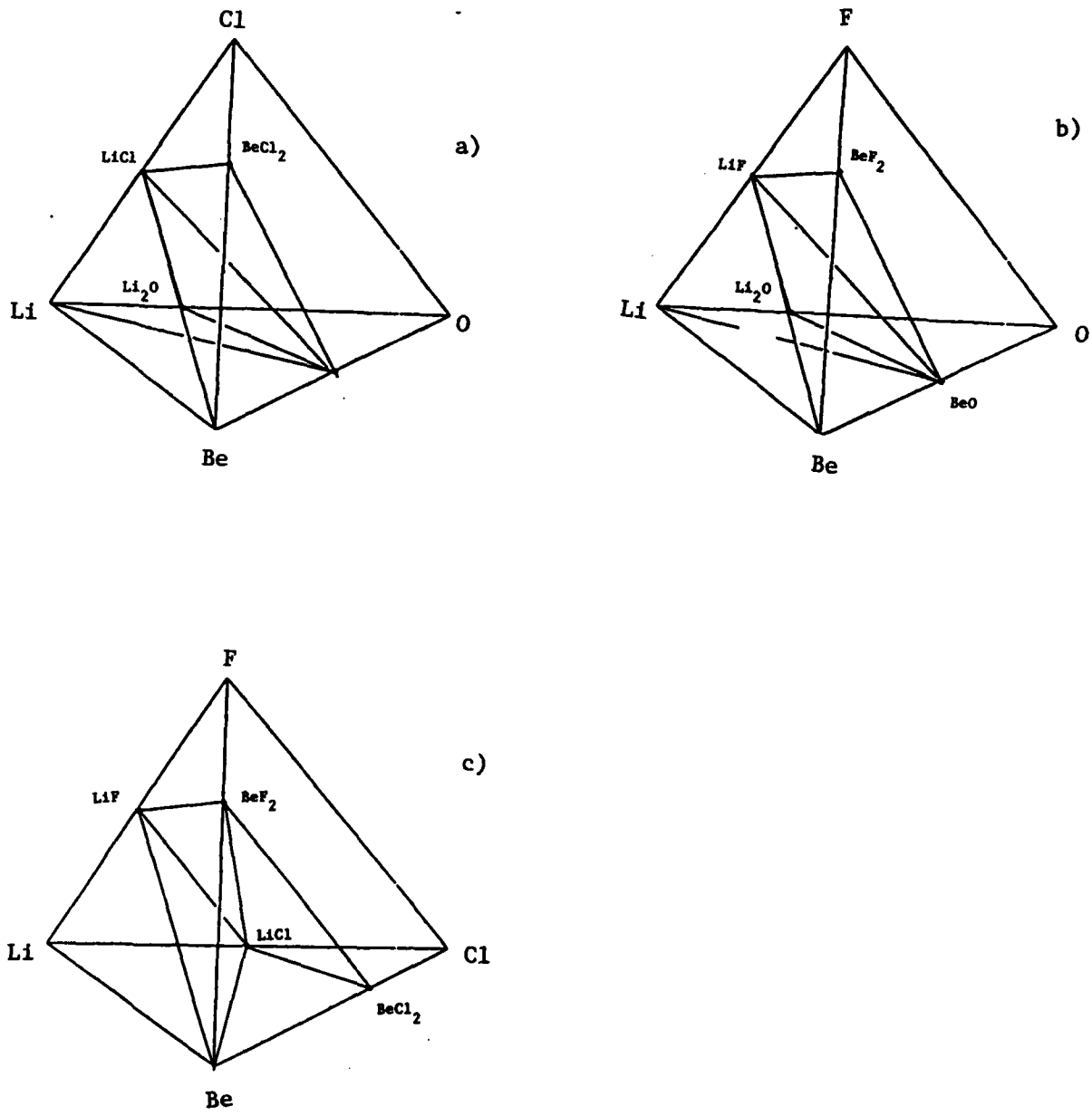

Figure 15. Outline of 1100 K, 1 atm phase compatibilities relating to oxidation of Li/Be mixtures by ClO_3F. (a) chlorination and oxygenation; (b) fluorination and oxygenation; (c) fluorination and chlorination.

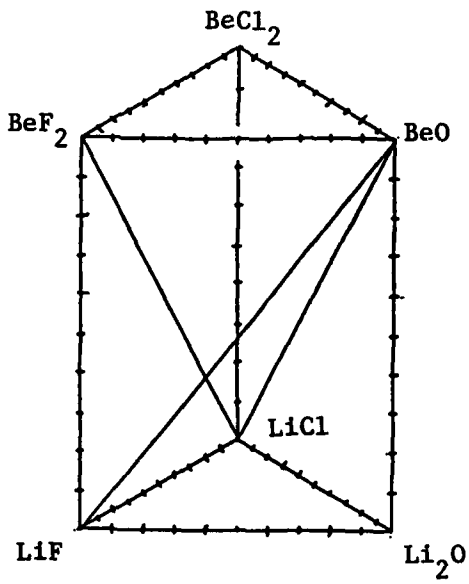

Figure 16. Outline of phase compatibilities among products of complete oxidation of Li/Be by ClO_3F at 1100 K, 1 atm.

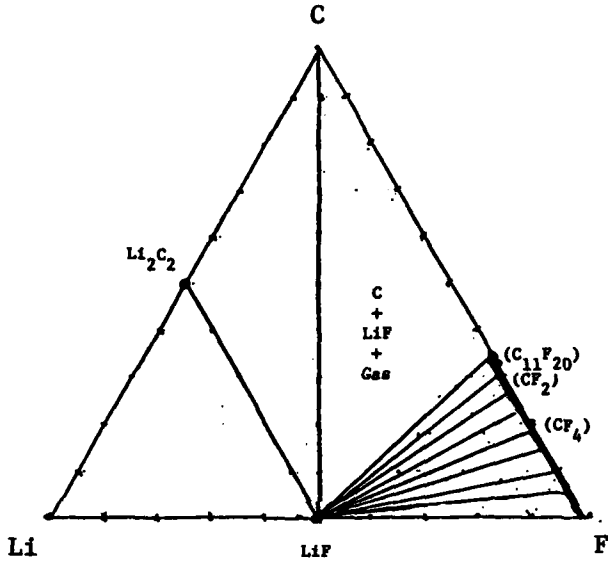

Figure 17. Calculated phase compatibilities in the system Li-C-F at 1100 K, 1 atm.

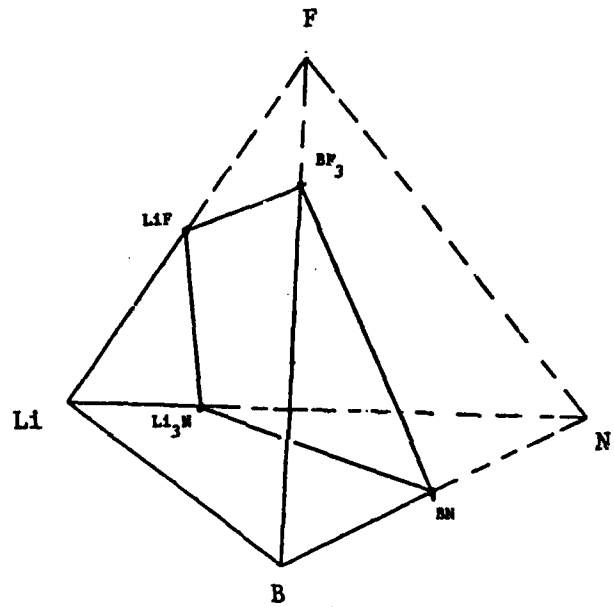

Figure 18. Quaternary reciprocal systems relevant to oxidation of LiB alloy by NF_3.

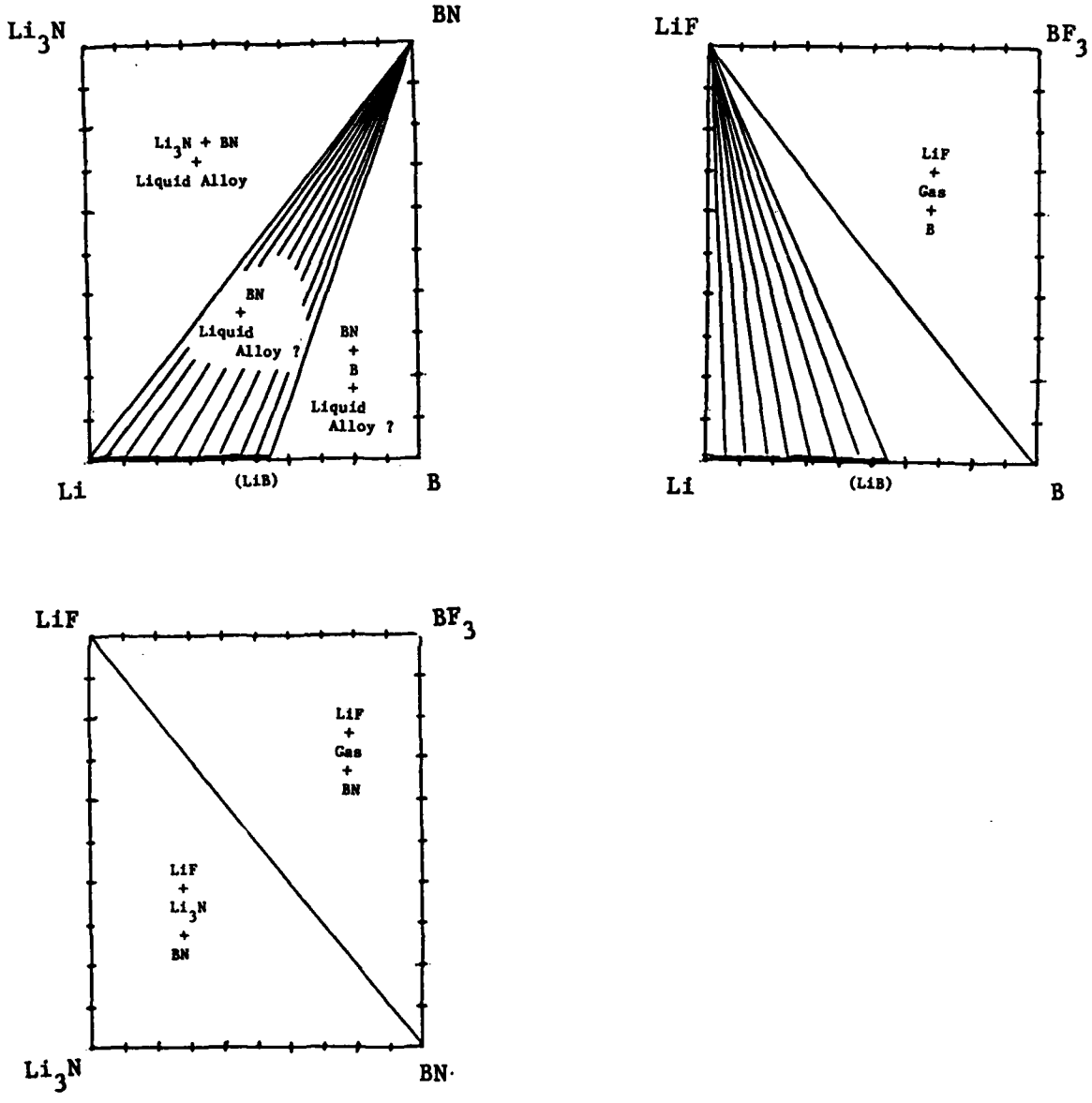

Figure 19. Calculated phase compatibilities in ternary reciprocal subsystems of the system Li-B-F-N at 1100 K, 1 atm.

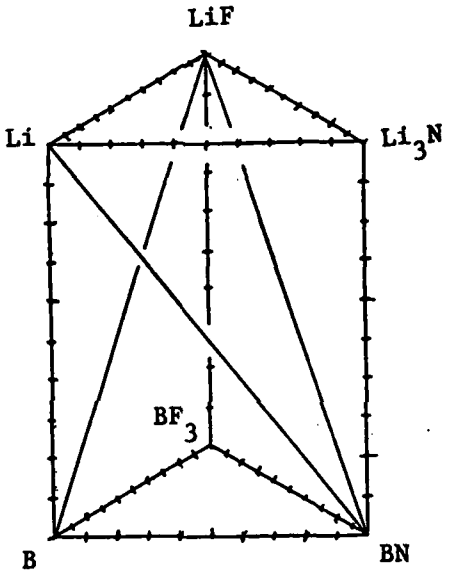

Figure 20. Outline of phase compatibilities among products of complete oxidation of LiB by NF_3 at 1100 K, 1 atm.

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11. ABSTRACT (A 200-word or less factual summary of most significant information. If document includes a significant bibliography or literature survey, mention it here)

A survey of eight alternative liquid metal stored chemical energy reactions has been made for purposes of comparison with the lithium-aluminum/water, lithium/sulfur hexafluoride, and other reaction schemes. The objective of the study was to survey the potential of these eight reactions as alternate stored chemical energy systems and to develop priorities for future study. Experimental data on the products of reaction and kinetics of reaction are presented for: (Li/H_2O; H_2/O_2), (Li/H_2O; NaO_2/H_2O; H_2/O_2), ($MgAl/H_2O$; H_2/O_2), and ($LiAl/ClO_3F$). These data have been collected using thermogravimetry and Knudsen effusion mass spectrometry, with x-ray diffraction analysis of experimental products. Among other results, the data show that the aluminum component of the fuels is relatively inert to oxidation up to 650 °C. Above this temperature, materials limitations have hampered the collection of experimental data.

Thermodynamic analysis has been used to extend the data on each of the eight reaction schemes, and to predict the chemical reaction with best represents the complete oxidation of each fuel by the indicated oxidant at 1100 K. Enthalpies have been calculated for each fuel/oxidant combination. Safety considerations are also discussed for each. Suggestions for future research are given, including suggestions for overcoming the materials problems encountered in this study.

12. KEY WORDS (Six to twelve entries; alphabetical order; capitalize only proper names; and separate key words by semicolons)
enthalpy yield, kinetics, liquid metal combustion, lithium, lithium-aluminum lithium -boron, lithium-beryllium, magnesium-alumium, nitrogen trifluoride, water

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